



INTERNATIONAL ATOMIC ENERGY AGENCY, VIENNA, 1968

## FAST REACTOR PHYSICS

VOL.I

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### PROCEEDINGS SERIES

# FAST REACTOR PHYSICS

#### PROCEEDINGS OF A SYMPOSIUM ON FAST REACTOR PHYSICS AND RELATED SAFETY PROBLEMS HELD BY THE INTERNATIONAL ATOMIC ENERGY AGENCY IN KARLSRUHE, 30 OCTOBER - 3 NOVEMBER, 1967

In two volumes

## VOL. I

INTERNATIONAL ATOMIC ENERGY AGENCY VIENNA, 1968

FAST REACTOR PHYSICS (Proceedings Series)

ABSTRACT. Proceedings of a Symposium organized by the IAEA and held in Karlsruhe, 30 October - 3 November 1967. The meeting was attended by 183 scientists from 23 countries and three international organizations.

Contents: (Vol.I) Review of national programmes (5 papers); Nuclear data for fast reactors (12 papers); Experimental methods (3 papers); Zoned systems (7 papers); Kinetics (7 papers). (Vol.II) Fast critical experiments (8 papers); Heterogeneity in fast critical experiments (5 papers); Fast power reactors (13 papers); Fast pulsed reactors (3 papers); Panel discussion.

Each paper is in its original language (50 English, 11 French and 3 Russian) and is preceded by an abstract in English with a second one in the original language if this is not English. Discussions are in English.

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#### FAST REACTOR PHYSICS IAEA, VIENNA, 1968 STI/PUB/165

#### FOREWORD

The Symposium on Fast Reactor Physics and Related Safety Problems, arranged by the International Atomic Energy Agency, was held at Karlsruhe from 30 October to 3 November, 1967. One hundred and eighty-three scientists, representing 23 different countries and three international organizations, attended. Sixty-three papers were presented.

It became clear that participants were confident that the fast reactor would become a major source of nuclear power and it was to be expected that the papers would show determined progress towards that goal. Three separate trends can be discerned.

First, much attention is being given to the heterogeneity problem and several different methods of calculation are being developed. Such techniques must be effective before it is possible to validate a cross-section set with the results of critical experiments and then to use it to do calculations, for, say, a power reactor with fuel in an entirely different physical form.

Second, several laboratories are using zone techniques to do effective fast-reactor experiments in thermal reactors. Although this calls for a high level of theoretical and experimental competence, it is well worthwhile, in view of the high cost of all-fast systems.

Third, much work, both theoretical and experimental, has been done in recent years on steam-cooled fast reactors, particularly in Europe. However, despite the apparent advantage of using heavy steam to reduce troublesome coolant void effects, very little attention appears to have been given to this point over the same period.

One of the most significant features of the meeting was the accumulating evidence that the parameter  $\alpha$  ( $\sigma_{capture}/\sigma$  fission) for <sup>239</sup>Pu in the 1-keV region was substantially higher than had hitherto been thought. This shift has serious economic implications for many fast-reactor designs at present under consideration, particularly those cooled by steam and those using oxide fuel. In fact, any plutonium-burning reactor might be less effective than previous calculations had led one to believe.

In view of the interest shown in this point, an impromptu panel discussion was organized on the last day of the symposium to discuss the new information. The group of experts considered three aspects of the problem: the best  $\alpha$  values to use; implications of the increased  $\alpha$  value; and ways of improving the existing knowledge. A record of this panel discussion is printed at the end of this book, which contains the full proceedings of the symposium.

For the success of the meeting thanks are due in large measure to the staff of the Kernforschungszentrum Karlsruhe, whose hospitality and help were greatly appreciated by all participants.

#### EDITORIAL NOTE

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For the sake of speed of publication the present Proceedings have been printed by composition typing and photo-offset lithography. Within the limitations imposed by this method, every effort has been made to maintain a high editorial standard; in particular, the units and symbols employed are to the fullest practicable extent those standardized or recommended by the competent international scientific bodies,

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## REVIEW OF NATIONAL PROGRAMMES

(Session I)

Chairman: B. I. SPINRAD

#### FEDERAL GERMAN FAST BREEDER PROGRAMME

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#### Abstract

FEDERAL GERMAN FAST BREEDER PROGRAMME. The Federal German Fast Breeder Programme started in 1960 at the Kernforschungszentrum Karlsruhe. The first approach was clearly scientific and concentrated on physics, safety and principal engineering studies. In particular, four major steps were taken in the experimental area: SUAK, STARK, SNEAK, and SEFOR.

After 6 years of hard and intensive work both sodium and steam were considered promising coolants of roughly the same overall potential. This led to the decision to pursue major engineering developments with these two coolants.

About 1969/1970 two prototypes of 300 MW(e), one for each coolant, will be built with the objective of demonstrating the breeding and electricity producing potential on a large and significant scale. The sodium prototype will be built by the Konsortium Siemens-Interatom, possibly together with Belgonucléaire of Belgium and the Dutch Neratoom. The steam-cooled prototype will be built by the group AEG/MAN/GHH. Thereafter it is intended to order two demonstration plants of 1000 MW(e) to meet the ultimate objective: to have 1000 MW(e) fast oxide breeder reactors available on a commercial and competitive basis by the end of the 1970s.

#### 1. THE GENERAL PROGRAMME

The Federal German fast breeder programme started in 1960, when the reactor physics group finished the design work for the Karlsruhe research reactor FR 2. The original approach was characterized by the idea to have not only a fast breeder but also a commercially competitive nuclear power station. This led us immediately into the more detailed examination of the fuel cycle, and our point of departure was then to study the oxide-fuel fast breeder concept. The physics of such a reactor is notably different from that of a metal-fuelled breeder. It was in particular the determination of power coefficients and, in this connection, the investigation of the safety features of such a reactor, which was of prime importance. In reflecting on the title of this meeting, we feel that it is still of prime importance. It was quite natural, therefore, that the first thing in Karlsruhe was the planning of a fairly large physics programme. From this evolved constantly increasing engineering studies. It was the choice of coolant that was the overriding problem. Attached to this was an experimental engineering programme. Alongside that we started a fuel-element development programme, which included the construction and operation of the necessary facilities, that is, apart from the FR 2. a number of hot cells and Pu fabrication facilities and others. At a somewhat later stage we emphasized even more strongly R & D work on facilities related to the fuel cycle. Euratom has been associated with this work since 1963. The present association will last until the end of

<sup>\*</sup> Work performed within the association in the field of fast reactors between the European Atomic Energy Community and Gesellschaft für Kernforschung mbH., Karlsruhe.

1967. In the course of the work it was possible to determine more specifically the target of the project: the Federal Republic of Germany wants to have 1000-MW(e) fast breeder power stations by 1980. These fast breeder power stations shall be competitive with the available lightwater reactors. The target for the fuel-cycle costs is 1 mill/kWh or less, and for the capital cost the target is \$US 100/kW.

To arrive at this goal it is necessary to have fast breeder prototype reactors. This class of reactor must be under construction by 1970 and be in operation by 1974. This gives enough time for one or even two parallel 1000-MW(e) demonstration plants. The successful operation of these plants will be the basis for the commercial phase of fast breeder reactors.

Detailed studies in Karlsruhe led to the conclusion that sodium and steam have the same industrial potential. But the quality of the potential differs. Sodium has the potential of a high gain breeder with the associated fuel cost benefit, and high gain breeding is ultimately, say beyond the year 2000, an absolute necessity. We still feel that capital cost is a problem, the industrial and commercial solution of which can be demonstrated only at a later date. On the other hand, steam-cooled breeder technology will directly evolve from present-day light-water technology with all the greatly increasing operating experience to come. Two 600-MW(e) lightwater reactor stations were ordered in the Federal Republic of Germany early in 1967 on a purely competitive basis without any subsidies whatsoever. A somewhat lower breeding ratio is associated with steam cooling, which partly compensates the capital cost advantage with respect to sodium plants.

In 1965/66 the important decision was taken to prepare the early construction of one sodium and one steam-cooled fast breeder 300-MW(e) prototype in the Federal Republic of Germany. At the same time it was decided that, instead of the national laboratory, industry must take the lead and the responsibility for these prototypes, because it is industry and not the government which shall handle the expected fast breeder market after 1980. Therefore, the Konsortium Siemens-Interatom will establish the sodium prototype, and the AEG/MAN/GHH group the steamcooled prototype. The Kernforschungszentrum Karlsruhe, as a National Laboratory, will have a double function: On one hand it will actively support the industrial work and accept certain tasks in the framework of the direct prototype work. On the other hand it will pursue a basic programme under its own responsibility that will broaden and strengthen the project work more generally. Therefore, the Fast Breeder Project is no longer a Karlsruhe affair, but a national effort to arrive independently at the competitive stage of reactors of the second generation i.e. the fast breeders.

Looking ahead, one realizes that the market for fast breeder reactors will be tremendous and at the same time it is clear that the Federal German national basis is somewhat narrow. During the last couple of years there has been an intensive contact with both Belgium and the Netherlands, as a result of which there will be a three-level or ultimately even a four-level, joint fast breeder venture. Belgian and Netherlands industrial groups, BelgoNucleaire and Neratoom, join the Siemens-Interatom Group, and the national laboratories of these countries, i.e. Mol and Petter, join Karlsruhe, and the three governments have a corresponding agreement which also provides the funds. There is hope that, at a later stage, the electrical ultilities of these countries will also come to a similar understanding. There is a good chance that Luxemburg will also join the group. A similar arrangement with other partners is under way for the steam-cooled breeder.

The Fast Breeder Project, which originated at Karlsruhe, is therefore a fairly large joint venture. The involved partners agreed to a fair partition of work. In so doing, the larger physics efforts remain at Karlsruhe, in co-operation with Mol and Petten. Bearing in mind that this meeting is concentrating more specifically on physics and safety this report concentrates on these topics.

#### 2. EXPERIMENTAL PHYSICS

As has been mentioned before, it was the investigation of the physics of an oxide-fuelled fast breeder with its low-energy neutron tail that attracted attention from the very beginning. In 1961 a four-step experimental programme was envisaged as is now described.

#### 2.1. SUAK

At Karlsruhe pulsed subcritical assembly SUAK makes use of the accumulated experience with 14-MeV pulsed neutrons. The fast assembly allows for decay measurements and spectrum determinations by the timeof-flight technique. For an easier interpretation of measurements only bare assemblies are constructed, which also contain as few materials as possible. So far three assemblies have been investigated. The first assembly consisted of pure uranium (20% enrichment) only. In the other assemblies different amounts of hydrogen-containing material were added to shift the neutrons to lower energies and to study the anisotropic scattering effect of hydrogen in greater detail. At the present stage of physics in Karlsruhe we are concentrating on, among other things, the effect of the interface between core and blanket. The spectrum in the core is fairly constant with respect to the radius, but in the blanket there are rapid changes of the spectrum with the radius (or z axis). The large mean free path lets this effect penetrate also into the core. The clean geometry of SUAK and the ease of experimental access will enable us to investigate these interface effects at SUAK in the near future. At present we have a joint French-German venture at SUAK, to compare exponential and pulsed experiments, also by using SUAK. This is the programme HEUREKA of the Cadarache project. It should be kept in mind that SUAK is not designed for the use of plutonium.

#### 2.2. STARK

The fast-thermal Argonaut reactor was the first critical assembly with fast neutrons on the European continent. The graphite column of the original Argonaut has been replaced by a fast core. Only enriched uranium can be used, but not plutonium. STARK was a very great help in developing the fast neutron experimental techniques at an early stage, in particular the spectrum measurement techniques. A pile 5

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oscillator for heated samples and a heated foil furnace for studies of the Doppler effect were also installed. Further, an investigation on pulsed sources, with particular stress on noise analysis techniques, was made. Noise analysis is a great help for determining the neutron lifetime and the reactivity features during shut-down including the approach to criticality. Alongside this, an analysis of coupled systems was carried out. STARK will continue to be important for preparing experiments of the larger and more complicated SNEAK machine.

#### 2.3. SNEAK

The fast zero power assembly is a full-scale critical facility designed in particular to perform large-scale Pu fuel experiments. It went critical on 15 December, 1966, the same date when the Cadarache facility MASURCA became critical. SNEAK is comparable not only with MASURCA, but with the United Kingdom zero power reactor ZEBRA, and the United States ZPPR in Idaho. Physics-oriented experiments are being conducted in the present phase with great emphasis on steam and the involved reactivity coefficients, and the mock-up of the two Federal German prototypes will come later. The measurements in SNEAK will ultimately give the physics reference points for the whole German-Belgian-Netherlands fast reactor project.

#### 2.4. SEFOR

The South-West Experimental Fast Oxide Reactor is a joint venture of General Electric and Karlsruhe together with Euratom, the South West Atomic Energy Associates, and the USAEC. As is well known, it is a 20-MW(th) experimental dynamic test reactor, the construction of which is fairly advanced. First criticality is expected for the early summer of 1968. The objective of this reactor experiment is to obtain information on the dynamic behaviour of oxide-fuelled fast breeders, particularly the Doppler coefficient, under operating conditions. At the same time engineering experience is acquired on a significant scale. The experimental programme provides for four distinct steps:

- (a) Static tests
- (c) Oscillator and balanced oscillator tests
- (c) Sub-prompt critical excursions
- (d) Prompt critical excursions.

At the recent Cadarache meeting there was a comprehensive paper on SEFOR that reported the latest stage of the work.

#### 2.5. KRITO

KRITO is at present a thermal critical facility at the Netherlands nuclear research centre at Petten. In the framework of the joint Karlsruhe-Mol-Petten programme Petten will transfer this facility into a coupled facility to investigate the reactivity influence of the fission products. More data on this will be reported by the Petten group at this symposium.

The reactor physics of fission products has been neglected for some time. This stems from the early days of a very hard spectrum and comparatively low burn-ups. But fast ceramic reactors of a high burn-up do have a considerable fission product poisoning, which decreases not only reactivity but also breeding with the result that fuel costs are again increased beyond a certain amount of burn-up as is the case with thermal reactors. Thermal reactors have their optimum around 30 000 MWd/t; large fast oxide reactors seem to have their optimum somewhere near 100 000 MWd/t. More precise information on fission product crosssections is necessary to give more accurate evaluations.

#### 2.6. Van de Graaff Accelerator

In Karlsruhe a 3-MeV Van de Graaff accelerator with a bunching facility is used for cross-section measurements in the fast breeder project. The experimental programme is concerned with the measurement of capture cross-sections of structural material and fission products in the energy range of 10 - 200 keV. Also fission cross-sections of the isotopes <sup>23</sup>9Pu, <sup>240</sup>Pu, and <sup>241</sup>Pu are measured. Further effort is under way for absolute measurements of cross-sections and neutron fluxes as well as for self-shielding factors and resonances in the keV energy region. It may be mentioned in this connection that cross-sections of shielding materials are studied in the MeV-range by the use of the Karlsruhe cyclotron.

#### 3. REACTOR THEORY

Reactor theory has always been of special interest at Karlsruhe. It was through reactor theory that Karlsruhe entered the field of fast reactors. Looking back one can recognize a number of phases in the development of fast reactor theory. Originally, one-dimensional diffusion theory with 11 or 13 groups was applied for calculating critical masses or enrichments, the spectra in core and blanket, and breeding ratios. Very soon the emphasis on the lower end of the spectrum led to a larger number of groups and the evaluation of energy self-shielding effects. There are only about 10% of all fissions below 10 keV where most power coefficients originate.  $S_N$  versions and other transport techniques were introduced somewhat later in the multigroup procedures.

The next stage was to concentrate on the calculation of the Doppler and coolant void effects. For this more extended perturbation codes had to be developed, and a detailed examination of the resonances of the neutron cross-sections was necessary. The more realistic evaluation of the sodium void coefficient necessitated two-dimensional calculations. In addition to the diffusion and  $S_N$  codes in two dimensions, groupcollapsing procedures had to be developed for this two-dimensional procedure. Multizone arrangements in core and blanket also required two-dimensional procedures. The preparation of self-shielded crosssections, including heterogenity-effects, one-dimensional calculations with numerous groups (say 26, 60 or more), group condensation in different regions of the reactor, two-dimensional calculations with a few-group (6 or so) perturbation theory, the calculation of reaction rates, the calculation of power coefficients and neutron lifetime, and the onedimensional burn-up code, have all been put together into one super-code. the Karlsruhe NUSYS system. This code has been the basis for all design work at Karlsruhe and the Federal German industrial groups during the past two years. Emphasis is now being put on the transport effects in the region of scattering resonances. Besides applying the well-known ELMOE code, a new technique using about 200 groups is being developed.

In a third stage attention is being concentrated on the non-idealized actual power reactor. The influence of fission products is being studied in detail. In addition, the influence and burn-up of control rods, twodimensional power distributions with control rods, the proper distribution of control rods, the influence of burn-up on the reactor behaviour, the change of the sodium void coefficient and breeding ratio with control-rod insertion and fuel burn-up, the change of power distribution with plutonium build-up in the blanket, the calculation of the different temperature coefficients in different regions of the reactor, and more engineering details, are being studied. We also envisage the large-scale application of Monte Carlo techniques in order, for example, to calculate streaming effects. These more elaborate and partially cumbersome procedures are mandatory for a responsible prototype design and describe to some extent the present stage of development at Karlsruhe. Fast reactor theory and calculation may arrive at its asymptotic stage if all these steps are combined into an even larger code, and ultimately we must look for three-dimensional calculations, when power stretching is required to reach full competitiveness. Explicit three-dimensional calculations are not available at Karlsruhe for more direct design of the prototype reactors. but they will be there in time to calculate best core-blanket management schemes and control-rod operation programmes. We are aware that an IBM 360/91 computer will be necessary for this, whereas for the prototype calculations described above, an IBM 360/65 computer can handle the supercodes exceeding NUSYS. Such machines will be installed at Karlsruhe next year.

#### 4. REACTOR SAFETY

Concern about reactor safety is as old as reactor theory and it is intimately related to it. Fast reactor safety is particularly challenging, because the configuration of an actual fast reactor is not that with the highest reactivity, contrary to the situation with thermal reactors. In the old days it was the melt-down accident which attracted the greatest attention. Loss of sodium reduced the reactivity of the original core configuration, and it was the slow core melt-down without additional neutrons, which destroyed the core and could have led to a second configuration of much higher reactivity and therefore to an extreme power excursion with subsequent production of large amounts of mechanical energy.

Large fast ceramic power reactors have a region with a positive coolant void coefficient in the inner part of the core. If the coolant is lost the production of neutrons is enhanced, and the destruction of the core is combined with the power excursion and the subsequent production of mechanical energy. Parallel to that one must now consider the large inventories of plutonium of these fast breeders, together with the previously unusual sizes of 1000 MW(e). This leads to new levels of

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radiological concern. We conclude that these hypothetical disasters produce mechanical energies of not much more than 1000 MWs, even if extreme measures to obtain a small sodium void reactivity effect like pancaking are not taken. The figure of 1000 MWs is a limit, because of the Doppler coefficient. In fact this seems to be the most important benefit arising from the Doppler coefficient. Rather sophisticated versions of the Bethe-Tait technique are now available at Karlsruhe, taking into account the cylindrical geometry and the axial structure of sub-assemblies; also considerable work is going on to determine the equation of state for the mixed oxide fuel.

It seems to be possible to contain 1000 MWs in a reactor building. Benefiting from the experience of SEFOR safety and licensing procedures, we arrived at the two-containment concept. There is an inner concrete containment around the reactor and the primary circuit; this is nitrogen filled and designed to withstand the 1000-MWs event, but has only a 10% tightness. Therefore a second containment, which is of high tightness and houses the rest of the reactor installation, is being planned.

The 1000-MWs incident can happen more or less only if the safety system fails completely. Measures of diversity and redundancy can avoid such a malfunction almost completely. There are a limited number of events like sodium voiding after a break in the primary circuit and subsequent melt-down, which cannot be affected by a functioning safety system. Therefore, a safe and reliable design of the primary circuit turns out to be as important as the design of the safety system itself. But we hope to arrive ultimately at a situation of engineered safeguards including that of the primary circuit. In such a case no reactor containment but an ordinary building is necessary. We are not yet confident enough to design such all-out engineered safeguards for the Na and steamcooled prototypes. Much more work on engineering components testing is required for that. To prepare this we started some time ago to work on the mathematics and statistics of reliability and availability, which is an exciting new area. In so doing we hope to identify the schemes and fields of excessive engineering testing thus ultimately arriving at the concept of engineered safeguards to be eventually applied to our 1000-MW(e) demonstration plants of about 1975/6.

In addition we will pursue several more obvious tasks connected with reactor safety like aerosol research, the radiochemistry of primary circuit contamination in the case of steam-cooled breeders, iodine filtering etc.

Fast reactor physics and safety is becoming an increasingly mature affair, which requires imaginative ventures with all the features of largescale scientific enterprise. But this will enable us to have a responsible prototype design by 1970.

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#### HÄFELE

#### DISCUSSION

M.F. TROYANOV: In this paper Mr. Häfele quite rightly stressed the need to know the fission product cross-sections in order to determine their effect on the neutron balance. He also mentioned the existence of an optimal value for the burn-up: what is this value, as obtained in these calculations?

H.W. KÜSTERS: As regards a 1000-MW(e) sodium-cooled reactor I can tell you that, on comparing the neutron and economic properties of the system, we arrived at a figure of about 70 000 MWd/t.

## FAST REACTOR RESEARCH AND DEVELOPMENT PROGRAMME IN JAPAN

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#### Abstract

FAST REACTOR RESEARCH AND DEVELOPMENT PROGRAMME IN JAPAN. The present paper reviews the fast reactor research and development programme, with the stress on fast reactor physics and related safety problems.

The first part of the paper deals with the research and development programme, which was prepared by a Special Committee for Power Reactor Development Project in compliance with the request of the Japan Atomic Energy Commission. The 100-MW(th) fast experimental reactor and fast prototype reactor are to be sodium-cooled. The former is scheduled to be critical in 1972, and some of the related R&D activities have already commenced. The prototype is to be developed at the latest by the early 1980s. A prototype advanced converter project is also now under way in Japan, and a new organization was established in October 1967 to develop both the fast reactor and the advanced converter.

The second part of the paper describes the research and development programme connected with fast reactor physics and safety problems related to the fast reactor project, and also outlines the present state of activities in Japan. The JAERI fast critical assembly, which attained criticality at the end of April 1967, is scheduled to play an important role in obtaining information on the reactivity effects in large fast reactors, and will be used to check designs of the experimental fast reactor and the prototype fast reactor. Also, other establishments plan to prepare additional experimental facilities for research on this subject. In many establishments theoretical research is increasing on the following topics: nuclear data for fast reactors, fast neutron energy spectrum, reactivity coefficients, design study of large fast reactors, analysis of fast reactor kinetics and accidents. The study of fast reactor programme is also being prepared on such subjects as containment system design studies, sodium technology and fuel development.

The annual rate of increase of energy consumption has become very great in Japan in recent years. However, our energy resources such as coal, oil and fissile materials are far from sufficient. For this reason, we are very interested in nuclear power generation, especially in efficient breeder power reactors. It is expected that sodium-cooled fast breeder reactors will be in use earlier than breeder reactors of other types.

The Japan Atomic Energy Commission (JAEC) has held, since 1964, the Power Reactor Development Conference where various specialists have participated to establish a power reactor development programme to establish an energy supply and to promote science and engineering in Japan. After the report was presented by the Power Reactor Development Conference in March 1966, the JAEC decided to establish a Special Committee for Power Reactor Development Project, June 1966. The Special Committee has been planning the national development programme for the fast breeder reactor and the advanced converter reactor, with the

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assistance of many subcommittees and working groups. A new organization, the Power Reactor and Nuclear Fuel Development Corporation will be established on 1 October, 1967, to promote the power reactor development programme with the aim of realizing the JAEC policies with regard to the fast breeder reactor and the advanced converter reactor on a wider basis. The organization will include the Japan Atomic Energy Research Institute (JAERI), the Atomic Fuel Corporation (AFC), manufacturing industries, electrical utility industries and universities.

To develop fast breeder reactors, the Special Committee recommends the construction of an experimental sodium-cooled prototype reactor in the near future. As a necessary step in this development, sodium technology has been studied since 1957.

The construction of the fast experimental reactor (JFER) is necessary in order to acquire experience in design, construction and operation of the reactor type by utilizing our research and development efforts and industrial technology. This experience will contribute to the design and construction of both the prototype fast reactor and larger reactors. The JFER will facilitate various irradiation studies with fast neutron flux.

The conceptual design of the JFER is almost completed. The prototype breeder reactor (about 200 MW(e)) will become critical around 1975 under the supervision of the new organization. The fast critical facility (FCA) will be used to carry out a mock-up experiment of the reactor core. The necessary theoretical study for the FCA and the reactor is also proceeding. A second design study of the 1000-MW(e) sodium-cooled fast reactor was completed recently.

With regard to the development of plutonium fuel, the main work has been done by the Plutonium Fuel Development Laboratory of the AFC. Close co-operation between JAERI and the new organization (including AFC) are to be maintained in the design, fabrication, construction and operation of these fast reactors.

#### 1. DEVELOPMENT PROGRAMME OF FAST REACTORS IN JAPAN

#### 1.1. Design of the Fast Experimental Reactor (JFER)

The 100-MW(th) fast experimental reactor (JFER) is scheduled to be under construction in 1969, and to go critical in 1972. The conceptual design is nearly completed including details of sodium coolant circuits and the various mechanisms of the components [1]. On the basis of the conceptual design, the kinetic behaviour of the reactor at normal operation and transient state are also being analysed. The necessary amount of <sup>239</sup>Pu investment is considered to be about 220 kg. It is expected to load mixed-oxide fuel of plutonium and uranium for the first fuel investment. Depending upon the availability of sufficient plutonium, uranium-oxide fuel can be loaded instead of mixed-oxide fuel. The cladding material of the mixed-oxide fuel pin will be AISI-316. The main parameters of the reactor are shown in Table I.

The 100-MW heat will be cooled by air at the first stage of reactor operation. After steady operation of the JFER is achieved, it can be used for testing the steam generator, if reliable technology for a steam generator is developed by then. The future installation of an in-core loop is being considered.

TABLE I. DESIGN FARAMETERS OF $100-MW(m)$	ETERS OF 100-MW(th) JFER
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Plant performance	
Reactor power (MW(th))	100
Core power (MW(th))	92
Reactor inlet temperature (°C)	350
Reactor outlet temperature (°C)	500
Core region coolant velocity (m/s)	6
Primary system total pressure drop (kg/cm <sup>2</sup> )	6
Dimensions	
Core volume (including control rods) (litre)	200
Pitch of fuel assemblies (triangular) (mm)	80
Pitch of fuel pins (mm)	7.7
Clad outer diam.(mm)	6.3
Clad thickness (mm)	0.35
Fuel diam. (mm)	5.5
Core height (mm)	600
Axial blanket height (mm)	400 × 2
Radial blanket height (mm)	1400
Gas reservoir height (mm)	200
Core performance	

Core average power density (MW/1)	0.46
Core material (enrichment)	PuO <sub>2</sub> - UO <sub>2</sub> (0.30)
Core volume fraction (unit fuel assembly) Fuel/Na/SUS = 38/38/23	
Core fuel density (g/cm <sup>3</sup> )	10
No. of core fuel assemblies	55
No. of control rods	12
No. of radial blanket assemblies	257
No. of fuel pins per core assembly	· 88
Max. fuel-pin linear heat flux (W/cm)	500
Peak-to-average power ratio in core	1.6
Max. heat flux (W/cm <sup>2</sup> )	250
Max. clad surface temperature (°C)	580
Max. fuel temperature (°C)	2300
Max. burn-up (at. %)	5

The reactor power will be obtained with the following considerations in mind:

- (a) Core height should be at least 600 mm in order to irradiate fuel elements for the prototype reactor in the JFER.
- (b) The best cylindrical geometry of the reactor core is desired in relation to an economic inventory of fissile fuel. The core volume is approximately 200 litres.
- (c) A moderate power density (0.5 MW/litre) in the core is required to obtain results of irradiation tests for fuel elements at shorter periods. Thus a moderate linear heat flux for fuel pin and neutron flux level is obtained, bearing in mind present fuel technology and future development.

The design principles of the JFER are generally conservative, since this is the first experience with fast reactors in Japan, and the safety considerations are paramount especially in Japan because of earthquakes and public concern. One of the features of the JFER is that the reactor is highly flexible for fuel testing. Since the reactor is not very large, the peak-to-average ratio of power rating in the core is inevitably not very low; however, the sodium coefficient is negative enough and the reactor is inherently safe. Long burn-up capability of the fuel is not necessarily required for the first inventory, and the minimum requirement is to provide stable irradiation conditions at the designed power level and thus to provide suitable fast neutron flux for fuel testing. Specially designed in-core monitoring systems are to be installed; also, an alphagamma monitoring cave capable of some non-destructive testing and the mechanical handling of fuel assemblies - e.g. the measurement of pressure drop along the fuel element by means of inert gas flow, and minor repairing of fuel assemblies removed from the reactor - will be constructed near the reactor. Periodic inspections of fuel assemblies at each stage will enable estimates of burn-up capability and safety for further irradiation to be made.

#### 1.2. 1000-MW(e) fast reactor and protype fast reactor

The construction of a prototype reactor will begin in 1972, and become critical in 1975. The 1000-MW(e) commercial reactor will have a 4- to 6-loop cooling system. If we choose a prototype with a 1- to 3-loop cooling system, the capacity of the prototype reactor should not be less than 200 MW(e), and 200 to 300 MW(e) is considered appropriate.

Design studies of the 1000-MW(e) fast breeder have been carried out by JAERI and will be followed up by the new organization. The first conceptual design study on the 1000-MW(e) fast breeder was completed in April 1966 [2]. The second conceptual design study was finished recently and will be published by JAERI. The results obtained by these design studies are reflected in the detail design of the JFER and in the design philosophy of the prototype breeder.

Two kinds of design study on the 1000-MW(e) breeder reactor were done by JAERI, based on the sodium-cooled, and mixed-oxide fuel systems. Safety, economy and breeding gain are three factors to be considered for such a commercial power reactor plant. Difficulties concerning the safety problems of the fast reactor increase as the reactor size becomes larger. The first design of the 1000-MW(e) breeder has a flat cylindrical

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(H/D = 2.15) core geometry in order to obtain a higher breeding gain (1.44). Two-zone loading of different enrichment (10.5% and 14.8% PuO<sub>2</sub>) is applied to get flat power distribution. The calculated sodium coefficient in the worst case is more than +3 dollars, while it could be less than +1 dollar for sodium loss in the central 19 channels.

A ring core geometry was selected in the second design study and safety aspects are emphasized because of the following points:

- (a) Maximum sodium coefficient is reduced so as to have a greater nuclear safety margin
- (b) Power density in the core is reduced so as to have a greater thermal safety margin
- (c) Clad thickness is increased so as to have greater mechanical strength.

#### 1.3. Fuel development

The detailed design of the fuel element for the JFER, hydraulic flow studies and mechanical assessments of the fuel assembly, are being carried out at JAERI. A JFER fuel assembly is being manufactured to study the coolant flow condition of the assembly in the water loop. Experimental data of flow pattern, pressure drop, velocity distribution and vibration are obtained, and the results are found to be in agreement with the expected values.

The development and studies of fuel material are mainly done by the Plutonium Fuel Development Laboratory in AFC [3]. Studies on cladding materials and on mixed carbide and nitride and some on mixed oxide are to be done by the metallurgy group in JAERI. Inspection techniques have been developed and are being applied in the Inspection Laboratory of AFC.

An irradiation test of fuel pins in the Enrico Fermi Reactor is planned in collaboration with JAERI, AFC and the Japan Central Research Institute of Electric Power Industry (CRIEPI). The fuel test assembly contains 24 fuel pins sub-assembled in four thimbles. The combination of sample pins is planned so as to obtain as much information as possible from the single test assembly irradiation.

Other series of irradiation tests are being planned in the Dounreay Fast Reactor. The first programme is to be performed with the main emphasis on the investigation into the cladding material. AISI 316 stainless-steel pipes of different grain size, cold drawn and solution treated, will be irradiated at a higher temperature in the reactor. Mixed-oxide fuel pins, the same as those designed for the first loading of JFER, are to be irradiated during the second stage of the programme.

Studies on mixed carbide and nitride are not yet started. However, JAERI is modifying research facilities for this purpose. Uranium carbide and nitride have been investigated during recent years by both JAERI and private companies. Interest is shown in the nitrocarbide.

#### 1.4. Sodium technology

Studies on sodium technology were started by Hitachi, Ltd., and JAERI participated a little later by sending staff. Several sodium loops with 1- to 3-in. piping and with heat generation up to 200 kW, were con-

structed by Hitachi, Ltd., and these were operated jointly by the two organizations during various studies on corrosion, impurity control, heat transfer and small-scale sodium component tests [4]. One of the loops was transferred to the JAERI site in 1965, and it has been operated to study sodium technology [5]. A sodium flow test of the JFER fuel assembly is also planned in this loop.

To establish the sodium technology for the development and construction of sodium-cooled fast reactors, a fairly large out-of-pile sodium test loop is now planned, based on experience to date. The heating capacity of the main heater, at the maximum temperature of the loop, is about 2 MW; the loop is therefore known as the 2-MW loop.

The primary purpose of this loop is to aid in examining various characteristics of the components for the experimental fast reactor that is to be built in the near future. It is also expected to supply valuable engineering data for the cooling systems of future large-scale fast breeder reactors. Domestic equipment and other facilities will be used for most of the loop system. Tests of the JFER fuel assemblies' characteristics are also to be performed by the loop.

Before the construction of the JFER, a mock-up of the main large components of the reactor will be fabricated and tested. Shielding plug, fuel-changing machine, control and safety rods, reactor vessel, dummy fuel assemblies and support plates will be assembled and tested in hightemperature sodium. The data will be used to improve the design, decide the accuracy in fabrication, and to prevent troubles during reactor operation.

#### 1.5. Reactor physics

The fast critical facility (FCA) is now completed at JAERI, which makes it possible to study the reactor physics of large fast cores for an extended period [6,7].

The machine is the split-table type of horizontal matrix structure. The matrix consists of 35 rows by 35 columns, but it is designed so as to be easily extended to 51 rows by 51 columns in which large cores up to 3000 litres with a 50-cm-thick blanket can be built. The facility is provided with a pulsed neutron source of 200 keV Cockcroft-Walton type accelerator, which generates neutron bursts from 0.05 to 10  $\mu$ s width at the centre of the assembly. The facility is housed in a double container with a 25-m diam. and height of 21 m. The facility became critical on 29 April 1967, with 20% enriched uranium. The experiment programme will be mainly concerned with the sodium void effect and the Doppler effect in large fast reactors. Accordingly, the heterogeneity effect in the sodium void experiment is now being theoretically studied. Regarding the Doppler experiment, an intermediate energy system, suitably moderated to enhance the Doppler effect, is proposed. The nuclear characteristics of the JFER core will be studied by the facility using plutonium fuel.

In the Atomic Energy Society of Japan, a Fast Reactor Committee was organized in 1964. Theoretical researches on fast reactor physics and feasibility studies on large fast reactors have been carried out, not only in JAERI but also in universities and in industries. The fast reactor safety analysis group of the Japan Nuclear Code Committee has developed several codes for the evaluation of fast reactor safety problems. Methods for measuring the various power coefficients of reactivity have been investigated. Other problems of interest are the improvement of experimental techniques, especially for spectrum measurements, and the development of noise techniques for the investigation into space-dependent kinetics. These techniques will be incorporated into the fast critical experiment.

#### 6. Fast reactor safety

With regard to safeguards in connection with the JFER, a 90-m-high meteorological tower will shortly be built at the proposed site and the observation of meteorological and diffusion parameters will commence. At present, emphasis is placed on developing diffusion and dilution analyses in calm conditions.

For the design of the primary containment of the JFER, a series of model experiments is scheduled in the near future to test blast resistance structure, top plug, blast shield, etc. As for the secondary containment, a study of the effect of sodium fire and a comparison between steel, reinforced concrete and pre-stressed concrete will be performed. An expperiment on sodium-air reaction is included in the programme for the near future. The aseismatic design of the containment system and reactor vessel and the study of shielding concrete are also important matters.

For research on the effect of sodium boiling at a transient condition on fast reactor safety, experiments using a water loop or a sodium boiling loop are now being planned. The fuel assembly plugging and melting accident and the counter methods against them are also to be investigated. In view of the importance of steam generator safety related to the prototype fast reactor, the experiment on water-sodium reaction will be started in the next fiscal year.

The theoretical safety analyses of the JFER are now under way; stability study, credible accidents, design and evaluation of control system, core meltdown accident and sodium air reaction.

## 2. REVIEWS OF FAST REACTOR PHYSICS AND RELATED SAFETY PROBLEMS

#### 2.1. Fast reactor physics

Studies on fast reactor physics have recently been in progress in Japan and the main results of the theoretical and experimental works were reported at the BNES London Conference in May 1966 [6]. Group constants, numerical methods, nuclear characteristics of fast systems, reactor kinetics, experimental techniques, pulsed neutron experiments and the fast critical facility (FCA), were reviewed with a list of references on work done since 1962 [7].

The core configuration of the FCA, which became critical 29 April, 1967 [8], is cylindrical. The core uses 20% enriched uranium and has a 30-cm blanket of natural uranium. The critical mass is estimated to be 96.7 kg ( $^{235}$ U), after an adjustment of excess reactivity of less than 0.1%  $\Delta$ k/k to the loading amount of 96.88 kg ( $^{235}$ U). The FCA was brought up to the maximum output of 100 W on 18 May, and the radiation doserate of the leakage from the FCA building was measured. The present

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core, slightly modified from the initial one, contains 97.62 kg, and the critical mass is estimated to be 96.3 kg. The calculated critical mass was 96 to 102 kg using the one-dimensional and two-dimensional diffusion codes and ANL-635 group constants.

An intermediate energy spectrum core programme has been proposed and will be carried out soon, particularly to obtain information on the group constants in the resonance energy region [9]. The intermediate core will be composed of 20% enriched uranium metal plates and a suitable amount of hydro-carbon compound or beryllium. As far as the resonance energy region is concerned, the intermediate core is expected to give the most sensitive experimental results. The experimental results obtained in such a system are not exactly the same as those obtained in a fast reactor system. However, useful information may be obtained on the cross-section set in the resonance region from the measured results of criticality, neutron spectrum, Doppler effect and other reactivity measurements.

The nuclear calculations of the JFER were made using the following codes: EXPANDA (one-dimensional multi-group diffusion) [10,11], TWENTY GRAND (two-dimensional few-group diffusion), TORCH (onedimensional burn-up), KPARAM (kinetics parameter) [12] ESELEM (neutron spectra, effective cross-sections, 1500 fine groups) [13]. These calculations for the criticality, reactivity coefficients, burn-up characteristics, control-rod worths and kinetics, etc. are contained in the progress report [1].

The sodium-loss reactivity effects of large Pu-U and Pu-Th fast reactors were studied [14]. Using the one-dimensional multi-group calculation, the maximum volume fraction of fuel for both fast systems were determined on the assumption that a positive reactivity effect is not caused by the total loss of sodium. It was found that the Pu-Th system has a much more negative reactivity effect than the Pu-U system upon the total loss of core sodium. The maximum fuel volume fraction is found to be about 5 vol.% larger for the Pu-Th system than for the Pu-U system.

The effect of fuel burn-up upon the breeding ratio, effective multiplication factor and sodium-void reactivity effect of large Pu-U fast reactors were investigated [15]. The initial breeding ratio may be a measure of the breeding ability of the fast reactor throughout its operational life. The variation of sodium-void reactivity owing to fuel burn-up of the above system is small.

A simple and quick method was proposed for one-dimensional multigroup calculations of multi-region fast reactors [16]. This method is based on the condition of neutron-balance in each of the regions or subregions. Neutron flux distributions are represented by the simplest functions. The accuracy is comparable with the ordinary diffusion theory calculations, while the computing time is saved by a factor of ten. The code using this method is called RNB (Regional Neutron Balance).

#### 2.2. Fast reactor safety

As part of the programme to develop the JFER, some research and development work on fast reactor safety has already started at JAERI. The following paragraphs outline the results obtained so far. An attempt has been made to derive a mathematical model by which the transfer function of fast reactors could be predicted accurately. A detailed distributed-parameter model was introduced, in which the temperatures of the fuel and coolant are evaluated as continuous radialaxial and axial functions, and also it was assumed that the expansion and deformation of the core structure depend not only on the temperature gradient of the coolant in its subassembly but also on the deformation of the upper structure. Being analysed with this model, the JFER does not show instability under credible conditions, and the increase of power and the decrease of coolant flow-rate tend to increase instability. Accidents such as various types of reactivity insertion, coolant coast-down and channel blockage are analysed using the code, EXCURS, which is somewhat modified from the original one developed at the University of Tokyo.

Dynamic analysis of the overall plant which contains the reactor, primary and secondary cooling systems and air cooler, the design and evaluation of the control system, and the determination of the operational method, have been made with an analogue computer. The control and operation system keeps the reactor inlet at constant temperature. For this purpose, the reactor inlet temperature is controlled by the control rod at constant power operation and at power-up the temperature is kept constant by decreasing the outlet sodium temperature of the air cooler as power increases [17]. To make a more detailed analysis, a hybrid system, which consists of an analogue computer, digital computer Pb 250 (Memory 8.9K) and independent simulaters of IHX (Intermediate Heat Exchanger) and the air cooler, is in the process of being formed.

In the field of hypothetical accidents, an analysis was made of the meltdown accident and the sodium-air reaction accident of the JFER. The AX-1 code was modified to calculate the explosion energy in the hypothetical melt-down accident more realistically. The main modifications are as follows:

- (a) It is modified to be able to take into account the Doppler feedback effect.
- (b) The insertion of the reactivity is given by the following equation:

$$k_{ex} = At^3 + Bt^2 + Ct + D + E exp(Ft)$$

(c) The pressure distribution of the core is given by the thresholdtype equation of state or by the saturation pressure-law equation of state.

The equation of state of the core materials is very important in calculating the explosion energy. However, the experimental data for the equation of state of uranium dioxide at high temperature are very poor. So it is calculated from the table of Hougen, Watson and Ragatz using the principle of the corresponding state. The code, EXPLO, with the above-mentioned modifications has contributed considerably in estimating the explosion energy and consequently in designing the primary reactor container.

A code was developed to analyse the sodium-air reaction. The main characteristics of the code are as follows:

- (a) The gas temperature is spatially uniform in the container;
- (b) The wall temperature is independent of place;

- (c) The heat generation rate by the sodium-air reaction is proportional to the concentration of the reactants and square root of the absolute gas temperature in the container;
- (d) Fission-product decay heat is taken into account.

The calculated results using the code have been applied to the design of the secondary reactor container.

The design and study of the containment for a fast breeder reactor system have also been made. Valuable experience was obtained through the construction of the containment system for the FCA at JAERI [18]. The necessary functions of the FCA containment were obtained by a double containment concept. The primary containment is to withstand the dynamic load due to the detonation. Apart from making a theoretical study. an experimental test was made, using the small-scale models and TNT explosives, to find empirical data for reinforced concrete for the primary containment. The secondary containment is a cylinder of reinforced concrete with a hemisphere cap. For the secondary containment, a comparative study was made for vessels of steel, reinforced concrete and pre-stressed concrete, from the economic and structural aspects. The aseismatic design is also an important problem because the foundation of the FCA building is on a surface sand layer far above shale rock. Much effort was centred on how the acceleration to the structure induced by earthquakes could be reduced to a minimum.

The fast reactor safety analysis group of the Japan Nuclear Code Committee at the Atomic Energy Society of Japan is developing fast reactor safety codes. This group is now preparing the computer code for the study of reactor transient due to coolant sodium boiling. The code is composed of two parts: thermohydrodynamics including coolant boiling, and reactor kinetics including the effect of various feed-back reactivity. In the thermohydrodynamics, a homogeneous two-phase flow model was used. The coding is almost finished and the transient induced by reactor power increase or plugging of coolant channel has been successfully traced for purposes concerned with reactor safety. In the case of a very rapid power increase, however, there still remains some difficulty in agreeing on the numerical solutions.

At the University of Tokyo, a study was made of the analysis of stability, accident and core meltdown of large fast reactors. For analysing the dynamic behaviour as a function of time, the code EXCURS was made [19]. The characteristics of this code are as follows:

- (a) The dynamic behaviour of the reactor is spatially independent and the energy group is treated as one group;
- (b) The thermal representation of the reactor is made by channel approximation. Two channels, hot channel and average, are taken in this code;
- (c) It is possible to vary coolant flow-rate and to take account of decay heat;
- (d) The Doppler effects, effects of fuel axial expansion, sodium expansion and structure expansion are included as feedback effect;

(e) It is impossible to analyse after fuel melting or sodium boiling.

An energy release during a core meltdown accident was examined for a typical large fast breeder reactor using the code FASTAC [20]. The FASTAC code is composed of a one-dimensional few-group diffusion equation and hydrodynamic equations, which govern the movement of the
core material. Axial movement of core material is represented by the change of axial buckling and the effective fuel density. The meltdown phenomenon is represented by the change of core height due to gravitational force and the corresponding change of core fuel density. These treatments are approximations of the original equations, but the calculated energy releases have shown good agreement with values previously calculated by other workers. The dependence of released energy upon the core volume was investigated and it was found that the linear relation exists between them when the change of the prompt neutron lifetime and the power-peaking factor are taken into consideration. The partial meltdown phenomenon was also analysed varying the spatial distribution of the initial temperature of fuel material. The available energy does not show a marked decrease irrespective of the initial meltdown area. On the other hand, when the Doppler effect exists properly the available energy is nearly proportional to the initial meltdown area and the meltdown is limited to the local part of the core according to the initial condition. No dangerous autocatalytic behaviour is found even in the case of the compaction of the fuel in the annular region of the core.

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# LE PROGRAMME FRANCAIS SUR LA PHYSIQUE DES REACTEURS A NEUTRONS RAPIDES

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#### Abstract — Résumé

THE FRENCH FAST REACTOR PHYSICS PROGRAMME. The paper reviews the work done by the Euratom-CEA Association in the field of fast reactor physics and discusses its place in the general programme.

Although RAPSODIE was not designed as a tool to be used in reactor physics, neutron measurements made with this reactor since it first went critical have shed considerable light on fast reactor physics in general and on the operation of this reactor in particular. To date, RAPSODIE has completed the equivalent of 70 operational days at the rated power of 20 MW(th); burn-up in the reactor core has now reached 9000 MWdA. Particular attention is being given to the problem of critical mass and to reactivity build-up, which can be an extremely valuable source of information on the processes taking place within the fuel.

Neutron studies in preparation for PHENIX, begun over two years ago, are now an integral part of the physics programme. They serve as the guiding thread for the Association's theoretical and experimental work.

The authors have access to the MASURCA critical assembly, the HARMONIE reactor, and the ERMINE fast-thermal critical assembly mounted within the MINERVE swimming-pool reactor. By kind permission of the Karlsruhe authorities, the authors can also participate in work on the SUAK sub-critical assembly.

The authors show how these four facilities where experiments are currently being performed on identical media of simple composition complement one another. The paper ends with an outline of current theoretical work and an indication of the underlying objectives.

LE PROGRAMME FRANCAIS SUR LA PHYSIQUE DES REACTEURS A NEUTRONS RAPIDES. Les activités menées par l'Association Euratom-CEA dans le domaine de la physique des réacteurs à neutrons rapides sont résumées, en les situant dans le cadre général du programme.

Bien que RAPSODIE n'ait pas été conçu comme un outil pour faire de la physique, les mesures d'ordre neutronique, faites sur RAPSODIE depuis la première divergence, ont apporté de nombreux et utiles enseignements aussi bien pour la connaissance de la physique des réacteurs rapides que pour la compréhension du fonctionnement de ce réacteur. RAPSODIE a accumulé à ce jour l'équivalent de 70 jours de marche à la puissance nominale de 20 MW(th), et le taux de combustion au centre du réacteur atteint maintenant 9000 MWjA. On aborde particulièrement le problème de la masse critique et celui de l'évolution de la réactivité, qui constitue une source extrêmement précieuse d'informations sur le comportement du combustible.

Les études neutroniques préparatoires à PHENIX ont commencé il y a plus de deux ans et elles constituent actuellement une activité essentielle dans le domaine de la physique. Elles constituent le fil conducteur des travaux expérimentaux et théoriques de l'Association.

Les moyens expérimentaux dont les auteurs disposent sont: l'assemblage critique MASURCA, le réacteur source HARMONIE et l'assemblage critique rapide-thermique ERMINE réalisé dans la pile piscine MINERVE. Grâce à l'obligeance de leurs collègues allemands, les auteurs effectuent également une expérience commune dans l'assemblage sous-critique SUAK de Karlsruhe.

On souligne la complémentarité de ces quatre installations dans lesquelles sont actuellement conduites des expériences sur un même milieu de composition simple. Enfin on indique les objectifs et les grandes lignes des études théoriques en cours.

Nous résumons ici nos activités dans le domaine de la physique avec l'intention de les situer dans le cadre général de notre programme sur les réacteurs à neutrons rapides. Comme on peut l'imaginer, notre objectif prioritaire consiste pour l'heure à faire fonctionner le réacteur RAPSODIE. Aussi commenceronsnous par parler des aspects les plus saillants de son comportement neutronique, et si nous insistons sur cette question, c'est parce qu'elle ne fera pas l'objet d'autres communications à ce colloque.

Bien que RAPSODIE n'ait pas été conçu comme un outil pour faire de la physique, il existe de profondes interactions entre ce réacteur et cette branche de la science; nous avons presque été surpris du nombre et de l'intérêt des enseignements que nous ont apportés les mesures d'ordre neutronique faites sur RAPSODIE depuis sa première divergence, ceci aussi bien pour la connaissance de la physique des réacteurs rapides que pour la compréhension du fonctionnement de ce réacteur. Signalons en passant que RAPSODIE fonctionne de façon très satisfaisante. Nous avons aujourd'hui accumulé l'équivalent de 70 jours de marche à la puissance nominale de 20 MW(th), dont 50 depuis le 1<sup>er</sup> septembre, début de la première période de marche en régime continu, et le taux de combustion au centre du réacteur atteint maintenant 9000 MWj/t.

Le premier résultat que nous avons obtenu est la valeur de la masse critique, toutes barres en haut, à une température du sodium uniforme de 150°C. Nous avons constaté par l'expérience une masse critique, ou encore un volume critique, inférieurs de près de 10% à ce qui était prévu. Sur le moment, cette différence nous a étonnés car les calculs avaient été faits en utilisant les résultats d'une expérience critique effectuée il y a trois ans sur l'assemblage critique ZPR-III d'Idaho. Les calculs ont ensuite été repris et l'écart entre la valeur prévue et la valeur mesurée a pu être expliqué, par une analyse très minutieuse des différences existant entre la maquette critique et le réacteur, chacun avec sa complexité propre. Nous en avons conclu qu'il est assez illusoire de prétendre simuler un réacteur par une maquette fidèle et que, d'une manière générale, il est préférable de réaliser des expériences plus fondamentales, dans des configurations plus pures et plus simples, conduisant à une interprétation plus facile et plus sûre, même si elles s'écartent délibérément de la structure du réacteur réel. Nous en avons également conclu que les physiciens auxquels incombe la tâche de prévoir la masse critique d'un réacteur n'ont pas seulement besoin de bonnes connaissances en physique des réacteurs; ils doivent aussi posséder les qualités de minutie qui font la force des comptables.

Passons sur les mesures relatives aux taux de réaction, aux indices de spectres, à la valeur des barres de contrôle, et insistons par contre sur celles qui concernent l'évolution de la réactivité et des coefficients de température et de puissance. La détermination de ces grandeurs, effectuée à plusieurs reprises depuis la première montée en puissance, constitue une source extrêmement précieuse d'informations sur le comportement du combustible au fur et à mesure que progresse son irradiation. Nous présenterons ici les résultats obtenus à ce jour, bien qu'ils soient encore préliminaires.

Tout d'abord, nous avons constaté, au cours des périodes de marche continue, que la réactivité diminue de façon tout à fait régulière et linéaire avec l'irradiation, à raison de  $-0,94 \cdot 10^{-4} \Delta k/k$  par JEPN (jour équivalent à puissance nominale) là où le calcul avait prédit  $-10^{-4} \Delta k/k$ .

Lorsque, à l'occasion des arrêts, on mesure la variation de la réactivité du réacteur à puissance nulle, on constate qu'elle évolue sans à-coup et diminue selon la même loi à raison de -0,94 ·  $10^{-4}\Delta k/k$  par JEPN.





Par contre, l'observation de l'évolution de la réactivité du réacteur à 20 MW(th) a fait apparaître un phénomène inattendu. Lorsque, après un arrêt, le réacteur est amené à nouveau à 20 MW(th), nous avons jusqu'ici constaté chaque fois un gain de l'ordre de  $10^{-4}\Delta k/k$ , toutes choses égales par ailleurs, par rapport à la situation du réacteur à 20 MW(th) juste avant l'arrêt. Cela apparaît plus clairement sur la figure 1. Ces gains apparents de réactivité ne sont que la manifestation de la diminution du coefficient de puissance. Cette diminution était attendue, mais pas de cette façon-là; elle se produit en fait par sauts à l'occasion des cyclages thermiques du combustible, et elle ne semble pas affecter la réactivité à puissance nulle.

Les mesures du coefficient de puissance, qui ont été faites au début du fonctionnement à pleine puissance (en juin 1967) et ont été répétées au cours de l'irradiation, en octobre, montrent que le coefficient de puissance a, dans son ensemble, diminué de près de moitié jusqu'à ce jour (fig. 2). Comme le montre le tableau, la composante liée au sodium et aux structures est restée pratiquement constante, alors que la composante liée au combustible a diminué de presque un facteur 3. Nous n'avons pas encore d'explication certaine des observations faites, qui traduisent une diminution progressive de l'allongement du combustible à chaque montée en puissance. Parmi les causes envisagées, citons:

a) le combustible oxyde se redistribue et se colle à la gaine qui, étant plus froide, freine la dilatation de la colonne;

b) la diminution du jeu entre gaine et oxyde diminue leur différence de température;

# TABLEAU. RAPSODIE - COEFFICIENTS DE PUISSANCE $(pcm/MW \text{ pour un débit de 800 m}^3/h)$

	Coefficient total	Combustible	Structure + sodium
Combustible se dilatant suivant la fibre la plus chaude	22, 5	14	8,5
Combustible lié à la gaine	15, 5	7	8,5

VALEURS CALCULEES

VALEURS MESUREES

Juin 1967	30,5	22	8,5
Octobre 1967	17,5	8	9,5

c) la disparition de la structure en pastilles entraîne que le combustible ne se dilate plus suivant la fibre la plus chaude, mais selon la température moyenne.

Nous n'insisterons pas davantage sur cette question, qui n'est pas encore complètement élucidée. Cela étant, nous nous attendons à ce que le coefficient de puissance diminue encore quelque peu jusqu'à une valeur analogue à celle qui a été calculée dans le cas d'un combustible lié à la gaine (soit environ -1,  $5 \cdot 10^{-4} \Delta k/k/MW$ ) et qu'il ne descendra en tout cas pas au-dessous de la valeur, de l'ordre de  $0, 9 \cdot 10^{-4} \Delta k/k/MW$ , qui correspond à la composante liée au sodium et aux structures. De toute façon il est clair que la mesure régulière de la réactivité et des coefficients de puissance et de température est de nature à apporter des renseignements extrêmement utiles sur le comportement moyen des 2300 aiguilles fissiles que contient le cœur au fur et à mesure que progresse l'irradiation.

Les résultats de ces mesures d'ordre neutronique seront ultérieurement confrontés avec ceux des examens faits sur le combustible irradié lui-même. Nous mettons au point en ce moment les méthodes d'analyse chimique et isotopique du combustible irradié, afin de comparer l'évolution réelle de sa composition à celle qui a été calculée.

L'analyse de l'évolution constatée sur RAPSODIE n'est évidemment pas directement transposable à des réacteurs de plus grande taille, dont la composition et le spectre sont notablement différents. Il est néanmoins certain que des effets analogues se produiront et nous en tenons compte d'ores et déjà dans nos études de PHENIX.

Les études neutroniques préparatoires à PHENIX ont commencé il y a plus de deux ans et elles constituent actuellement notre activité essentielle dans le domaine de la physique. Leur échelonnement est conditionné par deux dates clés: printemps 1968, où doit être établi l'avant-projet, et fin 1969, où doivent être fixées les spécifications pour la fabrication du combustible.

Pour nous guider dans ces études nous n'avons que très peu de références expérimentales car il n'a été effectué à ce jour dans le monde que très peu d'expériences intégrales au plutonium sur des milieux dont les compositions, les spectres, les teneurs en plutonium-240 soient voisins de ceux qui correspondent aux futurs prototypes. Nous devrons donc faire des expériences par nous-mêmes, et les moyens dont nous disposons à cet effet sont: l'assemblage critique MASURCA, le réacteur source HARMONIE et l'expérience critique rapide-thermique ERMINE.

Le réacteur source HARMONIE, d'une puissance de 2,5 kW, fonctionne depuis bientôt deux ans Il est consacré, pour la moitié de son temps, aux études de protection, l'autre moitié étant partagée entre la mise au point de techniques expérimentales et l'alimentation en neutrons d'assemblages exponentiels.

L'expérience critique rapide-thermique ERMINE, plus particulièrement destinée à la mesure de coefficients de réactivité, a démarré voici un an dans la pile piscine MINERVE située à Fontenay. Les premiers résultats obtenus et les caractéristiques de cette expérience feront l'objet d'une communication à ce colloque<sup>1</sup>.

<sup>&</sup>lt;sup>1</sup> BOUCHARD, J., VIDAL, R., MOUGNIOT, J.C., «Etude expérimentale des caractéristiques neutroniques de milieux multiplicateurs rapides dans l'assemblage critique thermique-rapide ERMINE», ces comptes rendus, SM-101/56.

Notre outil essentiel, l'assemblage critique MASURCA, est en service depuis 10 mois. Ses principales caractéristiques ainsi que quelques-uns des premiers résultats obtenus seront également présentés.

Le choix de nos méthodes de travail et du rôle dévolu à chacune des installations repose sur les remarques suivantes:

1) De nombreuses raisons s'opposent à une simulation aussi fidèle que possible du cœur de PHENIX. La plus évidente est la limitation de notre stock de plutonium - 180 kg environ, soit moins du quart de la quantité contenue dans PHENIX. La plus contraignante est sans doute la grande hétérogénéité des matériaux de simulation: vouloir respecter l'exacte proportion des noyaux constituants conduirait à des milieux très hétérogènes et n'ayant même pas un minimum de régularité.

2) La constituante d'un assemblage critique dans MASURCA demande plusieurs semaines ou même plusieurs mois, le coût de fonctionnement d'une telle installation est très élevé

En conclusion, nous n'étudierons dans MASURCA, en vue de l'étape PHENIX, qu'un petit nombre d'assemblages critiques, dont les caractéristiques seront choisies de façon à être aisément calculables, c'est-à-dire des cellules de composition simple et de petites dimensions.

Afin d'alléger au maximum la charge de MASURCA, les expériences qui y seront effectuées, et qui immobiliseront une grande quantité de matière fissile, seront consacrées à des évaluations précises du bilan de neutrons et à l'étude de la distribution spatiale de paramètres importants, tels que le coefficient sodium. La plus grande partie des études relatives aux propriétés « asymptotiques» des milieux choisis — indices de spectre, coefficients de réactivité, facteur de conversion, etc. seront menées, soit dans les assemblages sous-critiques associés à HARMONIE, soit dans ERMINE. On écartera également de MASURCA les mises au point de méthodes expérimentales.

Une telle répartition du travail n'est possible que si les résultats obtenus sur HARMONIE ou ERMINE sont parfaitement cohérents avec ceux qu'aurait donnés une expérience analogue effectuée dans MASURCA. Un recalage préalable était donc indispensable.

La première partie de notre programme a donc été consacrée, d'une part à évaluer les possibilités des différentes installations, d'autre part à effectuer ce recalage.

Nous avons choisi pour cela un milieu de composition particulièrement simple, le cœur 1-B de MASURCA, dont chaque cellule élémentaire est constituée par un barreau d'uranium enrichi à 30% et trois réglettes de graphite. Outre son extrême simplicité, ce milieu multiplicateur possède de nombreuses caractéristiques intéressantes:

- il présente un spectre suffisamment dégradé;

- sa masse critique est faible, ce qui a permis, dans sa version 1-A où chaque barreau d'uranium enrichi est remplacé par un barreau d'uranium naturel contenant 25% de plutonium, de réaliser au plutonium le cœur de démarrage de MASURCA;
- il permet une étude approfondie des effets d'hétérogénéité, grâce à la présence du graphite.

Conformément aux idées directrices que nous venons de rappeler, les propriétés du milieu 1-B de MASURCA seront étudiées dans quatre installations: l'assemblage critique MASURCA, l'expérience couplée rapide-thermique ERMINE, l'expérience exponentielle sur HARMONIE et des expériences pulsées dans l'assemblage sous-critique SUAK de Karlsruhe. Nous sommes heureux de pouvoir souligner, à cette occasion, l'excellente coopération qui a pu s'établir ainsi entre physiciens du KFK et du CEN de Cadarache. Cette première expérience commune a par exemple permis une confrontation fondamentale de certaines techniques en spectrométrie de neutrons rapides.

A MASURCA, ce programme se poursuit actuellement avec le remplacement progressif du cœur 1-A par le cœur 1-B. Les configurations intermédiaires sont destinées à nous donner le plus tôt possible une idée exacte des difficultés rencontrées lors de la réalisation d'expériences à plusieurs zones en uranium et plutonium et dans l'analyse des résultats obtenus.

Ce programme préliminaire se poursuivra jusque vers le milieu de l'année 1968. Durant cette période nous préparerons la suite en étudiant systématiquement, par le calcul, les caractéristiques des assemblages critiques à venir. Selon nos idées actuelles, ces assemblages critiques comprendront:

1) Deux milieux au plutonium, Z-1 et Z-2, destinés à l'étude des deux zones du cœur de PHENIX (Z-1 correspondant à la zone centrale la moins réactive). Pour définir ces deux milieux on recherchera le meilleur compromis entre le désir de se rapprocher de la composition exacte de PHENIX et le souci de la simplicité de réalisation à l'aide des matériaux de simulation de MASURCA.

2) Deux milieux à l'uranium-235, R-1 et R-2. Ces milieux, dits de référence, s'introduisent dans notre programme pour la raison suivante: le stock de plutonium disponible ne permet pas de rendre Z-2 – et a fortiori Z-1 – critique. Les montages critiques destinés à l'étude des milieux Z-1 et Z-2 comprendront donc nécessairement chacun une zone nourricière à l'uranium-235. Les milieux R-1 et R-2 constituant ces zones nourricières seront choisis afin d'être compatibles – selon des critères actuellement à l'étude – respectivement avec Z-1 et Z-2.

L'étude d'un recalage des spécifications de fabrication du combustible de PHENIX est envisagée par substitution d'une pré-série de combustible dans MASURCA. L'étude macroscopique de ces différents milieux dans MASURCA sera précédée et préparée par leur étude dans ERMINE ou HARMONIE selon le processus déjà employé pour les cœurs 1-A et 1-B.

L'exploitation des résultats de ce programme ne sera pas une tâche aisée, en particulier par suite de notre faible stock de plutonium. Néanmoins il n'est pas certain que ce soit forcément une mauvaise chose que de manquer de plutonium pour ces expériences, car cela oblige à un gros effort de réflexion pour les concevoir et les interpréter. Nous aurions tous raison d'être satisfaits s'il s'avérait possible de prédire avec une bonne précision les caractéristiques d'un grand réacteur - et nous pensons ici surtout aux réacteurs de 1000 MW(e) - sans avoir réalisé une maquette en vraie grandeur.

Parallèlement aux études menées spécifiquement pour PHENIX nous poursuivons un programme d'études à plus long terme dont le but est d'améliorer nos formulaires de calcul des réacteurs à neutrons rapides.

Nous nous contenterons de quelques très brèves remarques à leur sujet. Une première catégorie d'études vise à mieux calculer les diverses caractéristiques d'un réacteur à partir des données de base. Les sections efficaces dont nous disposons nous incitent à employer un très grand nombre

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de groupes; la complexité géométrique des réacteurs nous incite à découper l'espace en un très grand nombre de mailles. Il y a donc une tendance naturelle à augmenter la précision des calculs par l'emploi de codes de diffusion ou de transport de plus en plus complexes et faisant appel à toutes les mémoires que nous apporte chaque nouvel ordinateur. On peut se demander si de tels codes de calcul ne seront pas peu à peu supplantés par des codes de Monte-Carlo. D'autre part, il nous semble que dans bien des cas il y aurait intérêt à ne pas vouloir traiter le problème en une fois (en utilisant des codes qui prétendent tout faire), mais plutôt à essayer de développer des techniques de calcul qui permettent de calculer certains problèmes particuliers. Nous pensons ici spécialement aux bilans de neutrons en milieu infini, aux interfaces entre deux milieux, à l'hétérogénéité, aux barres de contrôle, etc. Nous savons que de nombreuses études ont déjà été poussées dans ce sens mais nous croyons qu'un effort supplémentaire devrait être fourni. Nous ne savons pas encore très bien comment les interfaces affectent la réactivité et les flux, et nous ne pouvons pas encore bien calculer le flux et le flux adjoint aux basses énergies; par conséquent nous ne savons pas encore bien calculer l'effet Doppler, ainsi que les coefficients de réactivité des divers matériaux.

L'étude séparée de certains problèmes particuliers permet beaucoup mieux qu'un calcul d'ensemble de bien comprendre la physique de certains phénomènes.

D'autres méthodes théoriques sont destinées à mieux exploiter les résultats des expériences intégrales. Il s'agira par exemple de déterminer la masse critique d'un réacteur de puissance sans pour autant recourir à une maquette critique fidèle et en vraie grandeur. Cet objectif ne peut être atteint que si nous parvenons à développer de bonnes techniques d'extrapolation. Nous savons tous que la théorie multigroupe, qui constitue une méthode de calcul universelle, n'est pas un bon outil d'extrapolation, et le devient de moins en moins au fur et à mesure qu'elle se développe en complexité et en précision. Nous ressentions tous le besoin de pouvoir représenter certaines caractéristiques essentielles de nos expériences et de nos réacteurs au moyen d'un nombre limité de paramètres sans pour autant sacrifier excessivement la précision.

Des efforts en ce sens sont poursuivis en France, mais la place nous manque pour en parler ici.

Nous voudrions seulement, en terminant, souligner que ce sont les impératifs du projet PHENIX qui guident et canalisent notre programme actuel sur la physique des réacteurs rapides, après que les études de conception de RAPSODIE eurent joué ce rôle pendant de nombreuses années.

On ne saurait trop insister sur l'intérêt, pour un programme consacré à la physique, de disposer d'un tel fil conducteur pour éviter que les études ne se dispersent et ne s'écartent trop des impératifs concrets.

#### DISCUSSION

J.L. ROWLANDS: Could we be told more about the error in the prediction of the critical size of RAPSODIE on the basis of the ZPR-III mockup experiments?

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What are the major differences in composition and geometry between the mock-up and RAPSODIE, and what are the problems of bookkeeping? Are there uncertainties arising from manufacturing tolerances?

F. STORRER: The basic differences between RAPSODIE and the critical experiment carried out several years previously on ZPR-III are as follows: Whereas RAPSODIE has an oxide blanket, the simulated blanket was metallic; sodium in the RAPSODIE core was simulated largely by aluminium; furthermore, there were appreciable fabrication differences between the pre-series and series assemblies and between these and the specifications. Thorough investigation showed that the details of geometry and composition had to be taken into account, particularly in the interface region between core and blanket.

In fact, since RAPSODIE has a  $k_{\infty}$  of close on 2, 50% of the neutrons produced in the core escape, and an error of 1% in evaluating these leakages gives rise to an error of 1% in the reactivity. These leaks should cause fewer problems for reactors now planned.

# UNITED KINGDOM PROGRAMME IN FAST REACTOR PHYSICS

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#### Abstract

UNITED KINGDOM PROGRAMME IN FAST REACTOR PHYSICS. The paper contains a review of the fast reactor physics programme being undertaken in the United Kingdom and describes the organization that directs the work.

#### 1. Introduction

There is a continuing programme of work directed towards the understanding of the physics of the Prototype Fast Reactor (P.F.R.) [1] and subsequent fast reactors. The data and methods used for making performance predictions are being revised to take account of new differential measurements and integral checks on sero-power reactors. This will produce more accurate predictions and, ultimately, will reduce the reliance on detailed mock-ups for the large fast reactors that will follow P.F.R.

The design calculations for P.F.E. have been performed using the data set, FD2 [2], derived from the U.K.A.E.A. Data Library. These predictions have been adjusted after comparison of experiments on the P.F.E. mook-up in Zebra with calculations using the methods used for reactor design. A new data set, FD3, is now being prepared from the current version of the Data Library, which includes the latest evaluated data. This set will be used to reinterpret a wide range of integral data obtained from the sero-power reactors, Zebra and Vera, and other reactors abroad. A considerable effort has been devoted to accurate measurements of neutron spectra so that such measurements can be included in a systematic attempt to adjust FD3 to suit the integral data within the limits of accuracy of the differential data.

This paper briefly surveys all aspects of the work in fast reactor physics in the U.K. It also describes the organisation within the U.K.A.E.A. which directs the work.

#### 2. Requirements of reactor design

In order to establish some of the main features of P.F.R., a parameter survey programme [3] was used. Additional exploratory calculations [4] were performed to decide other important features such as the optimum designs for the breeder blanket and the inner shield surrounding it. The likely features of subsequent, probably larger, reactors were also considered at an early stage so that as many as possible of these could be included in the P.F.E. When making predictions for the detailed design of the P.F.R., target accuracies were set for certain features. The value of k for a fresh charge of fuel (with the reactor at inlet temperature) was to be predicted within limits of  $\pm 2\%$ , in order to ease the problem of adjusting a fixed size of core to exact criticality. The power coefficient, predicted within limits of  $\pm 30\%$ , was sufficient to determine the kinetic response. Control rod worths, predicted within limits of  $\pm 10\%$ , were sufficient to decide the mass of absorber required per rod. Power distributions were to be predicted within  $\pm 5\%$  for a quite complex core and blanket configuration. The neutron and gamma fluxes at the nucleonic detectors [5] had to be predicted within such limits as would allow three ranges of instruments to cover the full range from shut-down to full power. The inner shield had to be designed to give acceptable levels of radioactivity in the sodium in the secondary coolant circuit.

A series of computer calculations was made to study various problems of fuel management for the operating reactor. The problems considered included several versions of the equilibrium fuel cycle and the approach to equilibrium.

The kinetic response of the reactor was studied for a range of flow and reactivity transients. A range of values for the individual contributions to the power coefficient was assumed, to allow for the uncertainty in predictions. It was then necessary to show that all events leading to damage to the reactor were extremely unlikely and that those leading to a district hazard were negligible. An analogue computer study of the whole power plant was also undertaken in order to facilitate the design of the control system.

Studies are being made of what happens in a sub-assembly in the event of total or partial blockage [6]. Possible ways of early detection of such events are being examined for example, by means of acoustic noise or electrical noise in thermocouples. In addition, idealised models of gross melt-down of the core are being considered to obtain an indication of the ultimate accident to guide the design of containment structures [7].

#### 3. Organisation and responsibilities

The engineering design of the P.F.R. is being undertaken by the Fast Reactor Design Office of The Reactor Group (T.R.G.) at Risley. Central Technical Services, T.R.G., Risley, deal with the immediate physics problems of the design office. They deal with performance calculations, fuel cycles, kinetics and accidents. They do the parameter surveys and economic studies using the "present worth" method [8]. Some theoretical methods are developed in C.T.S. and programmed for the computer.

Winfrith (T.R.G.) has the main responsibility for the development of physics design methods and the data to be used with them. The accuracy requirements for nuclear physics measurements are specified and the resulting differential data evaluated and converted into data sets suitable for the usual methods of calculation. Data and methods are checked and revised as a result of integral measurements made on Zebra and other

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zero-power reactors elsewhere. Theoretical assessment studies of advanced fast reactors with alternative fuels and coolants are undertaken. Research and development is also in progress on nuclear detectors, nucleonic instruments and control systems. The Winfrith shielding group is based at Harwell in order to make use of the reactor, Lido. This group recommends methods and data for shielding calculations, after checking them against experiments on Lido and Zebra.

At Aldermaston (Weapons Group), differential nuclear physics measurements are performed that yield nuclear data for fast reactor calculations. Many of the evaluations of the data are also carried out there. Integral experiments, including measurements of neutron spectra to check data and methods, are performed on the zero-power reactor Vera. The new reactor, Viper [9, 10], built for radiation experiments, gives information on the Doppler effect and promptcritical kinetics. At Harwell (Research Group), differential nuclear physics measurements are performed that yield nuclear data. Neutron spectra have been measured in sub-critical systems by time-of-flight spectrometry [11].

Dounreay (T.R.G.) is the site of the P.F.R. and so is specially concerned with the specification of experimental facilities and with operational problems. The Authority Health and Safety Branch at Risley, wets the safety aspects of the P.F.R. and is also developing Monte Carlo methods for use in safety assessments which are applicable to fast reactors. Some fast reactor calculations have also been performed by nuclear industry on contract to the Authority.

#### 4. Data and methods

The early calculations for the P.F.E. were performed using the data set produced by Yiftah, Okrent and Moldauer extended to 0.4eV by adding further groups. This 33-group set was called FD1 [12]. The long term aim was to use data whose origin and evaluation was better understood. The U.K.A.E.A. Data Library stores evaluated data at as many energy points as is justified. The computer programme, GALAXY [13], reduces the library to any chosen group arrangement. The set, FD2 [2], was the first produced in this way, in 1965, and has been used for all the detailed design calculations for P.F.R. This set is now known to lead to important errors in predictions [14] and new evaluated data are now available. A new set, FD3, is being tested.

For some years, there has been in existence a request list [15] that specifies the accuracies required in order to achieve, using differential nuclear data only,  $k_{\rm eff}$  for fast reactors to a standard deviation of 1%; breeding gain to a standard deviation of 0.03; and other properties needed in reactor design and in the interpretation of experiments. The measurement programme has gone some way toward meeting the objectives. Because of the difficulty of many of the outstanding measurements on the request list, it is expected that a successor to FD3 will ultimately be produced by adjustment of FD3 to suit integral data from sero-power reactors.

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Core calculations for the P.F.R. are usually performed with the multigroup diffusion theory code, CRAM [16], which is available in one-dimensional form in slab, cylindrical and spherical geometries and in two-dimensional form in RZ, R0 and XY co-ordinate systems. A two-dimensional triangular mesh version and a threedimensional version are under consideration. For one-dimensional calculations, the full group structure of FD2 can be used but, for two-dimensional cases, some compromise in groups and mesh is necessary. For example, the P.F.R. hexagonal sub-assemblies may be represented by rectangular somes with 4 mesh points per rectangle giving 1300 mesh points in a typical XY problem. This is solved with 6 energy groups to accuracy of convergence 0.0005 in  $k_{eff}$  in  $1\frac{1}{2}$  hours on the IBM 7090.

The self-shielding of resonance reactions in heavy elements in homogeneous media is allowed for by methods used by Abagyan et al [17]. Heterogeneous media are dealt with by equivalence relations. The effect on  $k_{eff}$  of the fine structure in the flux in zero-power reactors is calculated with the code, CELPERT [18], which solves the equations of integral transport theory in slab geometry. Both heterogeneity corrections are more important in the interpretation of experiments in zero-power reactors than for the more nearly homogeneous power reactors because of the thick fuel and diluent components used in the experiments, for economy reasons.

The worth of control rods is obtained by successive criticality calculations using CRAM, with the absorber in place and removed. This can only be done rigorously, with a two-dimensional computer code, at the core centre line. Semi-empirical extrapolations are used to yield the worth of off-centre rods.

First order multigroup perturbation theory is used for many calculations of temperature and power coefficients.

The accuracy of these methods has been tested by general studies and also for specific problems. In some cases, a correction has been applied to the result of the basic method. The studies made include: the effect of mesh size; the effect of number of groups in the group condensation; a comparison of diffusion theory with the  $S_N$  method of transport theory to various orders of N; and a comparison of first order and exact perturbation theory.

The approximate methods and equivalence relations used when treating the resonance region have been tested and revised as a result of more exact calculations using the codes GENEX and SDR [19]. The code, GENEX, produces data for heavy elements at close enough energy intervals to reproduce the resonances. The code, SDR, solves the slowing down equation in the resonance region exactly for a homogeneous medium and approximately, using collision probability methods, for multi-region systems. Up to 120,000 energy points may be used in the range below 25 keV and up to 8 regions.

In order to get reliable predictions for certain properties such as the sodium temperature coefficient, it is necessary to

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use a better group-averaging procedure than is normally used when deriving a data set like FD2. A development of the code, EIMOE [20], is being used for this purpose. It solves the fundamental mode equations in about 1500 fine energy groups. It is being developed as the regular intermediate step when producing a new data set from the Data Library.

A Monte Carlo code, GEM [21], developed for criticality problems, has been tested in exploratory calculations for the P.F.R. Special Monte Carlo methods [22] are being examined to calculate control rod worths in realistic three-dimensional geometry in acceptable computing times.

The calculations for the inner shield of P.F.R. were performed with a two-dimensional diffusion theory code, ATTOW [23]. A Monte Carlo code, MOB, was developed to treat the heterogeneity of this shield [24]. The theoretical methods were checked against experiments on Lido and Zebra [23, 24, 25]. Thetransport theory code, NIOBE [26], was used for calculations of the outer concrete biological shield for which diffusion theory is inadequate. Crosssection data reduced from the U.K.A.E.A. Data Library were used for most calculations.

#### 5. Integral checks on sero-power reactors; data adjustment

Fast critical assemblies are built on two sero-power reactors, Zebra at Winfrith, and Vera at Aldermaston. In both cases, the cores and blankets of interest are constructed of square plates, 3 mm thick, stacked in vertical steel tubes. This makes it possible to study a wide range of reactors with the same set of components. When a blanket about 30 cm thick is required, these reactors can accommodate cores of volumes up to 4000 and 200 1, respectively. The larger size of Zebra makes it more suitable for project work directed towards a particular power reactor; both can be used to yield integral checks of data or methods. Sub-critical assemblies at Harwell [11] and Aldermaston [27] have also been used for measurements of neutron spectra by the timeof-flight method.

Examples of the project work undertaken on Zebra are the studies on assembly 4 of possible moderating blankets for the P.F.R. [28]; on assembly 5 of the Doppler effect in the socalled Doppler loop [29]; and on assembly 7, the full-size The work on Zebra 7 includes studies of k P.F.R. mock-up. breeding gain, control rod worths, detector sensitivities and reaction rates in the core, blanket and inner shield. This work is complementary to more extensive studies of the inner shield on Lido [23]. The reactor, Dimple, has also been used in a thermal-fast soned critical arrangement to assist the conceptual design of a superoritical-steam-cooled fast reactor [30]. The reactor, Vera, has been used to study the effect of replacing the natural uranium blanket of the Dounreay Fast Reactor by nickel [31].

Experimental results from several reactors have been used to check the predictions using the FD2 set [14]. Some of the success of FD2 (and other sets) is due to compensation of errors 41

in Pu239 or U235 data that over-estimate k by errors in U238 that under-estimate it. An adequate test of a new set must cover a wide range of critical experiments, including plutonium-fuelled assemblies, few of which have been built to date.

Zebra is currently being used to extend the range of information on plutonium-fuelled systems. The value of G (capture-to-fission ratio) for Pu239 is being measured by the method used on Dimple [30]. A 15 MeV electron linac and 200 m flight path have recently been completed with which core and blanket spectra up to about 200 keV will be measured on the same systems by the time-of-flight method. The remaining parts of the spectra will be measured with hydrogen-filled proportional counters and Li6 spectrometers.

The programme for Vera is mainly concerned with measurements of simple assemblies for checking methods and data. A range of systems containing Pu239, U235, U238 and various diluents is being studied [32]. Recent work has included a homogeneous oxide assembly and further measurements on one of the earlier assemblies, 5A, in an attempt to improve the accuracy of the techniques of neutron spectrometry [27].

The new data set, FD3, is being produced from evaluated differential data; a successor to FD3 will be produced later, adjusted to force agreement between calculation and experiment<sup>\*</sup> for integral experiments. Work is in progress on the DUNDEE-PENICUIK system of computer codes [33] which may be used for this purpose. The work can be divided into three phases:

- Optimisation of neutron cross-section data adjustments to fit experimental critical sizes with no resonance self-shielding or heterogeneity effects (completed),
- (2) Optimisation of neutron cross-section data adjustments to fit experimental critical sises and spectra and spectral indices at the centre of systems with no resonance self-shielding or heterogeneity effects (well advanced),
- (3) As (2) but with resonance self-shielding and heterogeneity effects taken into account (planned).

As an alternative to the DUNDEE-PENICUIK system, it may be possible to use empirical adjustment methods which can take into account a wider range of integral data.

# 6. Integral checks on operating power reactors such as D.F.R.

The operation of the Dounreay Fast Reactor was reviewed in 1966 [34] when both physics and engineering aspects were considered. Because of its use as a fast flux test reactor, there is a requirement for accurate estimates of reaction rates including damage rates in graphite, steel and other materials. The methods used are similar to those used for the P.F.R. [31].

A simple analogue reactivity meter is proving itself a valuable operational tool in control rod calibrations, in worth measurements for monitoring kinetic stability and for exploring any suggestion of anomalous behaviour [35]. The reactivity noise is typically a tenth of a cent and step changes of a cent can be measured, for example, during worth measurements.

Noise measurement and correlation techniques have also been used in exploring reactor behaviour by recording on magnetic tape and extracting cross-and auto-correlations using off-line analogue equipment. The most interesting feature of this work has been the discovery of a flux oscillation at about 6 c/s whose frequency is proportional to reactor flow and whose relative amplitude is approximately independent of reactor power. This independence of power makes it unlikely that the oscillation could be due to a resonance in the power-induced feedback. The fluctuation has no operational interest but is being studied in case it should prove to be larger in a different design.

The general level of noise in a thermocouple has been monitored by an experimental piece of safety equipment. Typically, the part of the thermocouple noise signal, fluctuating at a frequency of about 0.15 c/s, is rectified and integrated for 5 min. It may be shown that, for a thermocouple underneath a sub-assembly (that is, downstream) the noise signal is a more sensitive indication of subassembly blockage than the steady signal.

The experience on D.F.R. has been used when specifying the experimental equipment and facilities for P.F.R. Study of the P.F.R. itself will be an important check on the design methods for subsequent reactors, especially for aspects like kinetics that cannot be studied on zero-power reactors.

#### 7. Discussion

There is a comprehensive programme of fast reactor physics in the U.K. Nuclear data is continually being measured, evaluated and incorporated into the data sets used for reactor calculations. Background knowledge, especially of plutonium-fuelled systems, will be extended by experiments on zero-power reactors. The integral data obtained will be used to adjust cross-section sets. Experiments will also be performed to study specific features of future reactor designs. Fast reactors are designed using multigroup methods which are now becoming well tried and tested against integral experiments and whose limitations are reasonably well understood. Work in progress will elucidate some of the remaining problems concerned with three-dimensional geometry, resonance shielding and heterogeneity. Studies are being made of fuel management problems and the optimising of future designs. Existing data and methods are adequate to design P.F.R. Work in progress will narrow the design uncertainties in subsequent reactors.

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# DISCUSSION

S. YIFTAH: You mentioned that integral measurements would be used to adjust the 35-multigroup cross-section set obtained by your group. Whenever a similar adjustment was made in the past using one set of integral experiments the results were good for similar systems but very poor whenever new, possibly bigger, systems had to be calculated and measurements compared. Supposing you adjust your 35-group set using integral measurements pertaining to the 250-MW(e) prototype, can you be sure that the adjusted system will be adequate for 1000 MW(e) or bigger reactors, or for predicting systems other than those used in the adjustment?

C.G. CAMPBELL: We shall adjust the new FD3 set to fit the results of integral experiments, but the integral experiments chosen will be those made in simple cell geometry under well-defined conditions. Integral experiments made on mock-ups may not be suitable. Consider, for example, the present knowledge of  $2^{38}$ U capture; there is a 15% uncertainty in the differential data. With present integral techniques the  $2^{38}$ U capture to  $2^{39}$ Pu fission ratio can be determined to 3% and may well be improved to  $1\frac{1}{2}$ %, but it is, of course, particularly important to examine all possible sources of systematic error in these integral measurements. It will be a long time before the accuracy of the differential measurement can approach that of the integral quantities measured with care in clean, welldefined lattices, and that allowance is made for the small spectral difference between systems, it should be valid to use these data in other situations.

H. W. KÜSTERS: As regards data fitting, our procedure was to take a group constant set and introduce certain changes, thereby producing corresponding changes in the integral quantities, which we then analysed in a similar, but simpler, way to that used by Mr. Pendlebury (Ref.[33] of the paper) so as to revert to the original set. The attempt failed. Would Mr. Campbell or Mr. Pendlebury care to comment on this?

C.G. CAMPBELL: One reason for your failure to return to the original data may be that you omitted to take account of the inevitable effect of the postulated change in data on the spectrum.

E. D. PENDLEBURY: In our optimization of group cross-section adjustments to fit experimental critical sizes we find that generally the data adjustments are in the direction implied by measurements taken more recently than those incorporated in the basic evaluations. This would indicate that the adjustments tend towards the physically correct crosssections.

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We have done calculations similar to those mentioned by Mr. Küsters and found the same effect, but this is readily understandable if one studies the mechanism of the adjustment procedure. For example, if the fission cross-section of <sup>235</sup> U, say, is the only quantity artificially changed, and then in the adjustment procedure all cross-sections are allowed to change. the adjustments will be spread over all the cross-sections and will not be concentrated on the <sup>235</sup>U fission cross-section alone, which means that the <sup>235</sup>U fission cross-section will in general not be adjusted to the same extent that it was artificially changed. It will, however, almost certainly be adjusted in the direction of the value from which it was changed. In other words, in the adjustment procedure, the group cross-sections are effectively assumed to be normally distributed about the starting values and hence the adjusted values depend to some extent on the starting values. Despite this effect, I firmly believe that cross-sections which have been adjusted to fit integral quantities are better than cross-sections which have not been so adjusted, provided the adjustments are reasonable in relation to cross-section measurements and their uncertainties.

R. VIDAL: I note that in the United Kingdom fast reactor physics programme a special effort is to be made to replace fission chambers by detectors. Has Mr. Campbell's group made comparisons of fission rates as measured by these two means?

C.G. CAMPBELL: Fission rate ratios are measured at Winfrith with foils of fissile materials placed in the appropriate plates of the lattice cell. After a standard irradiation, the induced fission product gamma-activity is measured and separate calibration experiments then relate the fission rate to the fission product gamma-activity. The calibration experiment uses absolute fission chambers in a back-to-back arrangement with a dummy chamber of the same geometry. A foil of composition appropriate to the chamber deposit is irradiated in the dummy chamber during measurement of the fission rate, then removed and the fission product gamma-activity counted. The spectrum is that of the lattice being tested although the same calibration factors are found (to within  $\pm 2\%$ ) even when  $^{235}$ U and  $^{239}$ Pu foils are irradiated in a thermal flux in which the fission cross-sections are known. We have found that the fission rate in the  $^{238}$ U chamber differs by 9% from that measured in the  $^{238}$ U fuel plates; this is why we use foils rather than chambers for this measurement.

# FAST REACTOR PHYSICS PROGRAMME OF THE UNITED STATES ATOMIC ENERGY COMMISSION

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#### Abstract

FAST REACTOR PHYSICS PROGRAMME OF THE UNITED STATES ATOMIC ENERGY COMMISSION. The United States of America fast reactor physics programme is a broadly based research and development effort that is focused on the major neutronics problems of fast reactor design and operation and aims at providing a sound basis for the large-scale commercial exploitation of fast power breeder reactors. These include the optimization of reactor performance to gain maximum economic advantages from fuel without sacrificing safety and reliability requirements. The major emphasis is on the physics of sodium-cooled systems with lesser effort currently on steam and gas-cooled systems.

The refinement of nuclear data for use in reactor calculations is a major high priority item. Such refinement will come from further specific differential cross-section measurements as well as correlations and/or adjustments via integral data and theory. The availability of large amounts of differential and integral data places severe requirements on automatic data processing techniques. These must be used both in the procurement and the compilation of the data to provide it in a form which is routinely useful for the reactor designer.

The continued operation of the fast critical facilities ZPR-III, VI, and XI, as well as the projected operation of the ZPPR, is planned. Much of the work on these facilities is in support of nuclear data verification of nuclear design methods. These facilities will also be used in support of engineering design of fast test and power reactors.

The development of reliable and relatively sophisticated nuclear design methods is extremely important so that the qualitative interpretive aspects of nuclear design can be minimized. The development of these methods requires that certain aspects of nuclear theory, in particular space-time kinetics, three-dimensional techniques, and/or variational methods must be applied to fast reactor systems. The safety analysis requirements include major accident and kinetics codes which provide for system transients.

Experimentally, the development of improved neutron spectrometry for fast reactors will provide a necessary basis for verifying theory and data for nuclear design. The full exploitation of the critical assemblies will give consideration to the use of automated data accumulation and processing, as well as improved techniques for reactivity measurements.

The Physics Programme is sensitive to the requirements of other disciplines in the liquid Metal Fast Breeder Reactor (LMFBR) Programme. These include the application of nuclear design methods to conceptual studies and safety analyses. Related quantitative studies and evaluation of the reactor fuel cycle are major considerations throughout the operating history of both conceptual and currently operating reactors. Such studies will provide feedback and specifications for the basic research and development programme.

# I. INTRODUCTION

The fast reactor physics programme of the United States Atomic Energy Commission is a broadly based research and development effort that concentrates on the major neutronics problems of fast reactor design and operation with the aim of providing a sound basis for the large-scale commercial exploitation of fast power breeder reactors. These problems include the optimization of reactor design to gain maximum economic advantages without sacrificing safety and reliability requirements. With the selection of the Liquid Metal Fast Breeder Reactor (LMFBR) system by the Commission and their decision to concentrate their major resources on the development of this concept, the main emphasis of the physics programme is in this area with lesser effort currently on steamand gas-cooled fast reactor systems.

The origins of the fast reactor physics programme in the United States of America date back to the very early days of nuclear energy and are intimately connected with early fast reactor feasibility experiments and demonstrations. The construction and operation of the Clementine fast reactor at Los Alamos in 1946 was an early effort to demonstrate some technical aspects of fast reactor technology. The completion and operation in 1951 of the first experimental breeder reactor. EBR-I. demonstrated the feasibility of operating a fast reactor system with a net breeding gain, and provided important information on operating stability and control. EBR-I operation started with  $^{235}$ U-fuelled cores and culminated with a core of plutonium fuel. The interest in fast breeder reactors for power production led directly to research and development in support of the second experimental breeder reactor, EBR-II. Having demonstrated the technical feasibility of breeding under power producing conditions, EBR-II is now being used primarily as a fast flux irradiation facility in support of our fuel development programme.

The Enrico Fermi Fast Breeder Reactor (FERMI) constructed at Monroe, Michigan, by Atomic Power Development Associates (APDA) and operated by the Power Reactor Development Company (PRDC) has likewise made significant contributions to establishing fast power reactor technology. The Los Alamos Molten Plutonium Reactor (LAMPRE) has contributed to the feasibility of operating fast reactor systems with molten plutonium fuels.

Many of the experimental techniques and analytical methods now in use in fast reactor physics applications were developed and tested as part of the original Manhattan Project. The thermal reactor development programmes and the intermediate spectrum reactor work have made significant contributions. For example, the  $S_N$  method used for transport calculations was one of several explored and developed at the Los Alamos Scientific Laboratory (LASL) long before the study and design of large fast breeder reactors, as we know them today, began. Simultaneously, the work leading to the routine application of multigroup methods was developed at the Knolls Atomic Power Laboratory. Many publications on this important method date back to the early 1950s. Similarly, early work on fast critical experiments was carried out at Los Alamos with the Topsy, Popsy, Jezebel, Godiva and related assemblies and provided data on the basic properties of reactors with very fast neutron spectra.

The early contributions to fast reactor physics from these fast reactor experiments, as well as other research and development not part of an integrated fast reactor programme, provided a valuable and substantial background upon which to base a future programme.

With the growth of the thermal power reactor industry and its potential of economic competitiveness with fossil fuels, the Atomic Energy Com-

mission (AEC) directed its attention to advanced concepts including fast breeder reactors as the next step in the utilization of the country's energy resources. The report "Civilian Nuclear Power - A Report to the President-1962" gave strong impetus to the organization of a national fast breeder reactor development programme in which fast reactor physics is an important basic ingredient.

Considerable study and effort was devoted to a survey of the status of the fast reactor physics work in the United States of America and other countries, and to identifying and analysing the technical problems that must be solved in support of the national fast breeder reactor programme. Also included in the survey were studies on how best to utilize the technical and industrial resources in the United States in accomplishing this work and in identifying the facilities which would be needed to serve as the "tools."

The studies resulted in a report<sup>1</sup> which identified a number of areas requiring the initiation of new work or the augmentation of existing efforts, and formed the basis for the AEC-supported research and development work during the past several years.

The aim of the research and development programme in reactor physics is to provide a sound basis for the large-scale commercial exploitation of fast breeder power reactors, beginning in the 1980s. The achievement of power at low cost requires large plant sizes (about 1000 MW(e) and optimum performance of all components. This will be approached by way of demonstration plants of lower power (about 300 MW(e)). These will be built and operated with less stringent technical requirements in the 1970s.

This two-step objective, defined by time, plant size, and graduated technical sophistication permits division of the research work into shortterm and long-term categories. The short-term objectives are characterized by requirements that ensure the development of a core design technology adequate for demonstration reactors in the early 1970s; and long-term objectives by the capability to produce, by the early 1980s, the high degree of economic optimization of reactor cores demanded for commercial applications without sacrificing safety requirements.

The LMFBR Program has already resulted in authorization to build or modify a number of facilities in support of programme objectives. These include the Oak Ridge Linear Accelerator (ORELA), the Argonne Fast Neutron Generator (FNG), the Zero Power Plutonium Reactor (ZPPR), the modifications to the Zero Power Reactors VI and IX (ZPR-VI and IX), and conversion of EBR-II to a fast flux irradiation facility.

The first reactor project arising out of the fast reactor programme is the design and construction of the Fast Flux Test Facility (FFTF) by the Pacific Northwest Laboratory (PNL). Committment to the design of the first demonstration power plant will probably be made by 1970. Readjustments to the physics programme to provide physics support to these projects will be made as needed.

The recognition of the need for planning for the extensive and difficult technical problems, which must be solved to achieve the goal of safe, economical and reliable fast breeder reactor power plants, led to the establishment of the LMFBR Program Office at Argonne National Laboratory (ANL). This office is staffed with technical experts in the various

<sup>&</sup>lt;sup>1</sup> Reactor Physics Efforts Required in Support of the Fast Breeder Development Program, WASH-1066.

disciplines for the purpose of giving technical advice and assistance to the Division of Reactor Development and Technology of the USAEC in the planning, co-ordination and evaluation of technical activities of the LMFBR Program. The Program Office is continuing the earlier physics review by incorporating the output of the various laboratories in the United States and abroad and by factoring in the technical input and requirements from United States industry. The Program Office is also developing the interactions between physics and other disciplines such as safety, control and instrumentation, plant systems, fuel cycles, fuels and materials, and core design. A revised and updated Fast Breeder Reactor Physics Program Plan will be issured during 1968.

# II. MAJOR FAST REACTOR DESIGN PROBLEMS

The major objective of the fast reactor physics programme is to develop reliable methods for nuclear design and to fully understand the neutronic behaviour of the reactor core under all operating conditions. The neutronic and related parameters of an LMFBR or other fast breeder reactor in either full-scale or demonstration systems cannot now be predicted within limits sufficiently well defined for optimum economical performance. Therefore, current and near-future reactor designs must be flexible enough to accommodate some degree of uncertainty even though a fairly detailed engineering mock-up is performed. Providing for such flexibility, and at the same time ensuring safe, reliable reactor performance, will prevent the attainment of the desired economical fuel utilization in the early designs. Ultimately, however, as better nuclear data and verified analytical techniques become available, increased confidence in predictive methods used for nuclear design should correct this deficiency.

The major physics problem areas and the objectives for reactors such as the Fast Flux Test Facility, an early demonstration plant, and a larger electrical capacity fast breeder reactor system are somewhat different.

The major problems for the Fast Flux Test Facility are primarily those associated with the design for maximum fast flux, the maintenance of constancy of neutron flux throughout reactor operating cycles, and the provision of flexibility to accommodate a wide variety of test irradiations. Some unique neutronic design problems are associated with the split conical core concept of the reference design. These will require extensive critical experiments for confirmation.

The demonstration and large-scale power reactors have problems associated with the management of fuel and with the large size of the cores. For the early reactors, reasonable prediction of criticality, the power distribution and changes of power distribution with fuel burn-up are needed. As one designs larger reactors, dynamics problems dealing with spatial effects and the potential influence of coolant reactivity in an abnormal situation become more pronounced than they are for the early plants. The ability to predict confidently the time-dependent reactivity of core configurations that might result from abnormal operations or malfunctions is required for safety analyses.

The understanding of the dynamic behaviour of large LMFBR reactors requires improved methods to describe the kinetics of such systems, particularly those systems having relatively weak neutronic coupling from one side of the core to the other. Also needed is a more accurate knowledge of the various feedback mechanisms and their effect upon the reactor operation. Similarly, the change in the interaction of relatively separate modular cores as a function of changes in the reactor core and blanket of each module with operation requires detailed study.

In the design of reactor control systems a number of problems arise, such as the specification of reactivity requirements, control rod mechanisms, composition, and the monitoring of the reactor flux. Self-shielding of control rods, while conceptually understood, needs to be explored for routine design considerations. The development of a reliable criticality meter for on-line computer control is needed. These are technical problem areas for physics and other design disciplines.

The design of optimum shielding has been and continues to be a problem for fast reactor designers. Shielding is not generally a question of conceptual feasibility; it is technically possible to shield practically any configuration. However, the economic optimization required for commercial plants makes it necessary to improve the accuracy of shielding calculations. Quantitative data on neutron and gamma interactions with structural components and clad materials in the reactor are needed to provide information on the ultimate component and clad lifetimes.

The burn-up of the fissile and fertile materials in the core and blanket, with the accompanying transmutations and fission products, produces changes in the interactions between the core and blanket. These changes result in altered criticality requirements, reactivity coefficients and power distributions which must be understood and be predictable. It should be noted that the approach to a routine or equilibrium operating fuel cycle will be strongly influenced by the isotopic content of the initial loading. Improved fuel burn-up codes for the precise prediction of reactor performance during core lifetime are needed.

The solution of major fast reactor design problems can be obtained in a number of ways; one is by demonstration in a high-flux high-temperature environment. The Southwest Experimental Fast Oxide Reactor (SEFOR), currently under construction by the General Electric Company, is an example of such a reactor experiment. This project is an example of international co-operation in the physics programme; it is also a major technical undertaking in the fast reactor programme as a precursor to a larger power reactor. The physics measurements that will follow reactor start-up and operation at power are expected to provide valuable demonstrations of steady state and transient feedback parameters, particularly the effective Doppler reactivity, and contribute substantially to the confidence with which the physics problems are understood and solved in the design.

# III. THE FAST REACTOR PHYSICS PROGRAMME

The major parts of the current and future fast reactor physics programme include: (1) Cross-section measurements, sensitivity studies, compilations, and evaluations, (2) neutron spectrum measurements, (3) critical experiments mainly on the Zero Power Reactors (ZPR-III, VI, IX, and ZPPR), (4) development of theoretical methods for nuclear design, and (5) interdisciplinary work including that to help guide subsequent physics research and development.

# A. Cross-section measurements

A significant part of the United States fast breeder physics programme is devoted to basic neutron cross-section measurements. There are three major efforts in this programme devoted primarily to meeting LMFBR cross-section requirements. The facilities utilized or to be utilized by these efforts are: (1) The Rensselaer Polytechnic Institute (RPI) Linac; (2) the Oak Ridge National Laboratory (ORNL) 3.0-, 5.5-, and 7.5-MeV Van de Graaff generators and electron linear accelerator (ORELA): and (3) the ANL 3.0-MeV Van de Graaff generator and 8-MeV Fast Neutron Generator (FNG). In addition to these efforts, there are several other United States measurement programmes, not primarily devoted to meeting LMFBR requirements, that have provided significant amounts of data useful to reactor designers in the past and are expected to provide further contributions of this type to reactor technology in the future. These include, among others, the work of LASL using large, underground, single pulse sources; work at universities such as Columbia and Duke; work by industrial groups such as General Atomic and Texas Nuclear Corporations; and also work in other physics programmes at ORNL, Brookhaven National Laboratory (BNL), ANL, and Livermore Radiation Laboratory (LRL).

The work under way at RPI at present is concentrated on capture cross-section measurements on a wide variety of materials, including separated isotopes of Fe, Ni, W, Pb, Al and Na. Measurements under way by ORNL, in co-operation with RPI, include the Pu capture/fission ratio (alpha at less than 1 keV). ORNL is also performing measurements of the ratio of  $^{238}$ U capture  $^{235}$ U fission in the energy range 0.1-1 MeV, and the elastic and inelastic scattering cross-sections for Fe and Na in the energy range >4 MeV. At ANL elastic and inelastic cross-section measurements are being made on a wide variety of structural and control materials, in the energy range 0.3-1.5 MeV.

At present a significant effort is devoted to the completion of the ORELA and FNG facilities and the development of special data-handling and analysis systems. The ORELA, for example, is scheduled for routine operation in September 1968 and the ANL Fast Neutron Generator (FNG) for operation in September 1969. A data acquisition system for ORELA is being assembled and efforts are under way to complete the specifications of an on-line immediate analysis system with light pen and display capabilities for use with the ORELA. An immediate data-analysis system complete with light-pen capabilities has recently been developed for use with the ORNL Van de Graaff generators and has greatly reduced the time required for the analysis of elastic and inelastic scattering data obtained with these facilities. A data link was recently completed and tested between RPI and the New York University (NYU) CDC-6600 computer. This is expected to greatly speed the analysis of the capture cross-section data at RPI and enable earlier utilization of these data by the reactor designers.

With regard to future plans, attempts will be made to obtain much higher resolution and accuracy in the energy range below 300 keV. Simultaneous measurements of more than one parameter will be emphasized, e.g., total capture, scattering, fission. In the 2-10-MeV energy range,

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elastic and inelastic cross-sections will extend the work at lower energies on fast reactor materials. The plans are best summarized by stating that they are based upon making those measurements, which can be made with the listed facilities, to meet those fast breeder requests listed in EANDC 103 "U", the Compilation of Requests for Nuclear Cross-Section Measurements (June 1967). The highest priority measurements will generally receive the earliest emphasis. In the next five years, an intensive cross-section measurement effort is envisioned.

# B. Sensitivity studies

It is difficult to determine precisely what the value is of any single measurement to the development of a fast breeder economy, since definitive answers to this question can easily cost more than making the measurements themselves. Nonetheless, the question is often asked by the measurers. Some attempts have been made to answer this question in terms of available information, experience with various "evaluated" cross-section libraries, via analyses of the anticipated FBR design problems, and in terms of the alternative methods which would rely more heavily upon critical experiments and integral methods. Some studies of this type were reported at the Neutron Cross-Section Technology Conference in March 1966 (See USAEC CONF-660303, Books 1 and 2). Other studies will be completed in time for the forthcoming conference on Neutron Cross-Section Technology (Washington, March 1968). Information in this area also will be generated in the data testing programme of the CSEWG, discussed below.

# C. Compilation and evaluation

The large quantities of cross-section data points being generated by the various groups mentioned above have produced a host of problems for the reactor physicists who utilize these data. They have been forced to develop automated methods for data storage, retrieval, evaluation, and the production of multigroup libraries. A few years ago, the Sigma Center Information Storage and Retrieval System I (SCISRS-I) format was developed by BNL for storage of experimental neutron cross-section data. The SCISRS-I tape now contains about a million data points.

A cross-section evaluation programme was developed utilizing experienced and capable personnel available at a variety of United States reactor organizations. This programme, co-ordinated by the AEC and the Cross-Section Evaluation Center (CSEC) at BNL, has culminated in the release of the Evaluated Nuclear Data File-B (ENDF/B) library, which was developed by the Cross-Section Evaluation Working Group (CSEWG). Similar efforts will be a continuing part of the future programme and will be effectively carried out through the National Neutron Cross-Section Center recently established at BNL.

Some 50 isotopes are included in the ENDF/B library, which will now undergo extensive testing via calculations of the characteristics of critical assemblies and relatively simple configurations for which reliable integral data are available. Codes have also been developed for operations upon ENDF/B and for converting ENDF/B to multigroup formats for use in the design and analysis codes of the various United States reactor organizations. Write-ups on the evaluations now in ENDF/B and the associated codes are also nearly complete. Plans include the development of a SCISRS-II data storage system which will eliminate some of the shortcomings of SCISRS-I and permit wider use of it. The ENDF/B tapes will be revised to form a second version (after perhaps a year of testing) and an evaluated cross-section library for shiel-ding will be developed which will be compatible with the ENDF/B system now available.

Significant progress has been made in the area of automated evaluations. A programme called SCORE has been developed by Atomics International (AI), using IBM facilities at Palo Alto, to display data from SCISRS-I or other files, such as ENDF/B, utilizing a large computer on a time-sharing basis. This permits a very efficient interaction between an evaluator and the great mass of cross-section data, which is stored on magnetic tapes. Analysis via light-pen overlay techniques and curve-fitting routines is thus greatly aided. Version I of the SCORE programme is completed and is being prepared for general release. Plans in this area include further development of curve fitting techniques and the incorporation of multilevel fitting procedures into the SCORE system.

#### D. Neutron spectrum measurements

Efforts at ANL continue to improve the proton recoil spectrometer described at the 1966 ANL conference. Measurements are made routinely in the ZPR criticals using this proton recoil spectrometer. Similar measurements are now being made by AI in the Epithermal Critical Experiment Laboratory (ECEL) to assist in the analysis of the Doppler measurements made at this facility.

The use of time-of-flight techniques for spectrum measurements is being carried out at General Atomic (GA) and RPI using relatively simple assemblies of those materials expected to be utilized in breeder reactors. The electron Linacs at these sites are used to generate fast neutron pulses within these assemblies. The early emphasis in these programmes has been on the development and calibration of detectors and the development of suitable data-handling and data-analysis systems. Recent measurements of fast neutron spectra from large assemblies of Fe and  $^{238}$ U have been reported by RPI. Measurements on small, spherical assemblies of  $^{235}$ U and  $^{238}$ U have been reported by GA.

Time-of-flight studies with simple systems, in addition to further developing the experimental technique, will also assist in the analysis of basic cross-sections and calculational techniques. Comparative measurements using the proton recoil spectrometer and the time-of-flight method, especially in the lower energy overlap region are planned during the next year. Provision is also being made for a time-of-flight capability at the ZPR-VI and IX facilities using the ANL Fast Neutron Generator to provide the necessary pulsed neutron source.

# E. Critical assemblies

The largest experimental effort in the United States fast breeder reactor physics programme relates to procuring integral data using fast critical assemblies. These data are needed not only to help develop and check analytical methods, but to verify actual core designs through the performance of mock-up criticals. For these purposes, the AEC has four major facilities at its disposal, utilizes or supports other industrialowned facilities, and has under review the question concerning whether additional facilities may be needed.

The bulk of the fast reactor physics critical experiment programme has been and will continue to be performed in the four government-owned facilities ZPR-III, ZPR-VI, ZPR-IX and the latest and largest ZPPR. The many experiments and mock-up criticals that have been performed in the past on these critical assemblies are well described in the literature. The Proceedings of the International Conference on Fast Critical Experiments and Their Analysis held at ANL in October 1966 include a good summary of the results up to that date. Some of the current and future activities will be described below.

First, in support of our efforts to develop analytical methods and techniques, ZPR-III is engaged in a series of basic, clean plutonium criticals with a broad range of spectra intended to bracket the spectrum range of interest to fast power breeders. These criticals are deliberately made simple to facilitate analytical correlation. During these experiments, the standard critical data are being procured including neutron spectra (using both proton-recoil spectrometry and foil activation techniques), critical mass, flux mapping, fuel and material worths, Doppler effects, sodium void effects, and heterogeneity studies. This series will be interrupted this autumn to permit an extensive programme of criticals to be performed in support of the Fast Flux Test Reactor (FTR) design. The recent basic investigations, identified as Assemblies 48, 48B, 49, and 50, developed general data for plutonium fuelled systems with differing spectra by substituting coolant in the reference configuration (48), with void (49), and with graphite (50). Assembly 48B was used to explore <sup>240</sup>Pu effects.

ZPR-VI has recently been engaged in extensive studies of the characteristics of a large-scale (2500-litre core) uranium carbide fuelled system. These studies are important not only in themselves for the information they provide on the properties and characteristics of carbidefuelled fast reactor cores, but in particular because they are an integral part of a programme conducted in both ZPR-VI and ZPR-IX designed to test the validity of using zone techniques to reproduce the characteristics of much larger reactor cores. Early results indicate that meaningful measurements can be made in reasonably small zones. The importance of verifying the validity and limitations of zoned-core measurements is obvious in optimizing the utilization of our limited and expensive plutonium critical fuel supplies.

ZPR-VI will soon be used to study an oxide fuelled simulation with  $\sim 3700$ -litre core volume. The zoned loadings in ZPR-IX have been used for Doppler reactivity investigations with a variety of neutron spectra. These were adjusted by varying the ratio of  $^{238}$ U to carbon from  $\sim 0$  to 1.4 in a reference composition (Coolant/structure/uranium/carbon = 35, 15, 25, 16 vol.%, respectively). Other recent data include importance measurements with californium sources to aid in the interpretation of zoned configurations.

As the planning of our integrated fast reactor physics programme developed it became obvious that insufficient plutonium critical facilities existed to carry out the programme. Accordingly, it was decided to modify the ZPR-VI and ZPR-IX facilities to permit them to conduct experiments with plutonium loadings up to 1000 kg in size. It is expected that these two critical assemblies will be available for plutonium experiments by this time next year.

As the concepts for the large-scale LMFBR designs developed, it became obvious that none of the existing facilities would be capable of handling the large mock-ups necessary for the large-scale power reactors. After considerable evaluation and review it was decided that a large wafer-type critical of basically the same design as ZPR-III, VI, and IX would best serve our needs and give us both the size required and the flexibility for interchanging our limited plutonium fuel supply between the four facilities and would permit intercomparison of results, not only with our own criticals, but with those of other countries. Accordingly, the Zero Power Plutonium Reactor (ZPPR) has been authorized and is under construction. This facility will be available for plutonium experiments by mid-1968. It is planned first to perform some large-scale basic experiments to check out techniques on large cores where the spectrum is soft and the signals achieved from perturbation methods are expected to be much reduced.

The magnitude of the fuel programme involved in "stocking" this reactor with fuel and non-fissile materials may be of some interest. In order to be able to analyse with some degree of precision the integral data developed from our plutonium criticals, we have insisted that all the fuel of a given plutonium composition be blended to close uniformity. The bulk of the fuel (2900 kg) will contain 11.5% <sup>240</sup>Pu, but a few hundred kilo-grams of 30% <sup>240</sup>Pu fuel will also be produced to investigate higher isotope effects and several hundred kilograms of rodded fuel will be produced so that rodded zones can be studied.

As the fast breeder reactors become larger and the neutron spectra become softer, it is essential that all aspects of the heterogeneous differences between wafer-type criticals and rodded-type reactors be thoroughly understood. Extensive heterogeneity studies will be conducted to insure that the critical experiments performed in the wafer-type criticals can be interpreted properly for the pin-type hot sodium reactors that will be built. Included in these studies will be experiments utilizing zones of plutonium pins both at ambient and elevated temperatures initially in ZPR-VI/IX facility. Many other studies involving various geometric groupings and drawer alignments of fissile, fertile, structural and coolant materials, will be performed.

For several years Doppler measurements have been made at Atomics International (AI) using the ECEL on a wide variety of fuels and structural materials in spectra ranging from a few kilo-electron volts to several hundred kilo-electron volts median fission energies. Recent measurements have been completed using Pu samples having various isotopic ratios. Also under study by AI is the effect upon Doppler measurements of samples of materials which are placed in fuelled zones whose temperature can be varied.

BNL is gradually shifting the emphasis of its work from thermal reactor studies to fast reactors. A Fast Source (FS) Reactor has recently been completed and will be used for the development of experimental techniques and special studies, such as the effects of heterogeneity and neutron streaming upon the interpretation and application of plate-type critical measurements.

# F. Theory and computational methods

A major United States effort in this area is the development of the Argonne Reactor Computation, the ARC, system, which is being developed on the IBM System 360 Model 50-75 computer complex. This system links a wide variety of computational modules and control programmes so they can operate in a unified manner without human intervention. For a detailed description of this system reference is made to USAEC Report ANL-7332, which will be issued in the near future. In recent months progress has been made in the development of: burn-up and depletion modules, codes to study heterogeneity effects, codes to compute effective fission and capture cross-sections in the resonance region, codes to interpret Doppler measurements, cross-section modules based on the Multigroup Constants Code (MC<sup>2</sup>), and certain transport and diffusion modules.

Additional computational efforts include: the development of burn-up codes at LASL and Babcock and Wilcox (B & W), space-time dependent kinetics studies at Massachusetts Institute of Technology (MIT), LASL, ANL, and BNL, development of Monte Carlo methods at AI and ANL, and sodium void analyses at APDA. The codes ETOE, ETOM, GAND, ETOG, and ENDRUN have been developed for utilizing ENDF/B data in the various industrial design codes.

Several analytical areas are receiving increasing attention in the United States of America by a number of organizations. These include proper averaging of differential data to produce multigroup parameters, flux synthesis methods and problems related to core melting patterns during large power transients. The BISYN code recently developed at GE is an early attempt to apply synthesis methods to fast reactor analysis and early results are encouraging. Included in the cross-section averaging efforts are those to rapidly and routinely adjust multigroup parameters for energy and spatial self-shielding by expanding upon the work of Bondarenko.

It is anticipated that future efforts will be devoted to the improvement of numerical and statistical methods for handling the transport of neutrons. With the new generation of computers, it is planned to exploit Monte Carlo techniques, for example, in the calculation of reactivity changes caused by changes in temperature or localized removal of coolants. The further development of two- and three-dimensional codes based on transport, diffusion and Monte Carlo methods is anticipated for the solution of burnup, control, and space-time kinetics problems.

# G. Interdisciplinary activities

Many applied aspects of the physics research and development require involvement with other disciplines in the LMFBR Program. The 1000-MW(e) LMFBR study programme, commonly referred to as the "1000 MW(e) Follow-Ons," is being pursued by the Atomics International, Babcock and Wilcox, Combustion Engineering, General Electric and Westinghouse Electric Corporations in co-operation with Argonne National Laboratory. These studies require an evaluation of the physics methods used to define the characteristic neutronic behaviour of various LMFBR concepts. The results of these studies will provide useful guidance to physics research and development. Another aspect of interdisciplinary activities is the development of codes that couple the various disciplines. Recent developments include the coupled neutronics, heat transfer, and hydraulics code, FORE-II, by General Electric and the quasi-static excursion code, QX-I, by Argonne National Laboratory. Related code development is being carried out by Atomics International and Pacific Northwest Laboratory. PNL, in conjunction with APDA, is refining some major accident codes based on AX-I, RAC and the Weak Explosion Codes for safety analyses. Related to these is development of codes to account for fuel slumping during excursions as input to the major accident codes.

#### SUMMARY

While at various places in this paper references are made to technical accomplishments, these are not presented in detail and are given more in the nature of programme progress reporting.

The technical accomplishments of the United States fast reactor physics programme are rather widely published. In addition to reports and papers in the technical and professional society journals, broad distributions is made throughout the world by the AEC Technical Information services. The Proceedings of the International Conference on Fast Critical Experiments and Their Analysis (October, 1966, Argonne National Laboratory), and the Proceedings of the British Nuclear Energy Society (BNES) (May 1966) are compilations containing summaries of the United States fast reactor physics results. The Proceedings of the Conference on Neutron Cross-section Technology (CONF-660303, Book 1 and 2, Washington, D. C., March 1966), and the Topical Meetings of the American Nuclear Society (Fast Reactor Technology, San Francisco, 1967; Coupled Reactor Kinetics, College Station, 1967; and Mathematics and Computation, Mexico City, 1967) are further sources of recent developments.

In the preparation of this paper we have discussed the reactor physics programme as though it were limited to the activities and efforts of United States scientists. We have not referred directly or indirectly to the contributions from scientists in other countries. We all know that much technical information and many ideas pertinent to fast breeder reactor physics have crossed the borders of the originating countries. The United States appreciates the considerable information it has received from the programmes of other countries. Indeed we believe that the area of reactor physics is an appropriate one in which the experience developed in the various countries should be freely exchanged. We look forward to continued co-operation and exchanges of information.
### GENERAL DISCUSSION

#### Review of the USSR fast reactor programme<sup>1</sup>

N.V. KRASNOYAROV: I propose to give a brief review of the USSR programme for fast sodium reactors. You will of course know that, on the basis of experience acquired with the fast sodium reactor BR-5 and in designing larger reactors, we have begun to develop the BN-350 atomic power station employing a fast neutron reactor of 1000 MW(th) capacity. Since the characteristics of this reactor are given in the literature, I will not dwell on them here.

The various components of the reactor are still at the trial stage, but construction of the reactor building at Shevchenko is nearing completion and assembly of the equipment has begun. The physical characteristics of the full-scale reactor model with a loading of approximately 1100 kg of  $^{235}$ U were tested in the BFS critical assembly.

The subsequent development of higher-pressure reactors with sodium temperatures capable of supplying steam to modern turbines, high burnup and long intervals between shut-downs for refuelling, has given rise to a number of problems. It is intended that the BOR-60 reactor, now under construction, will clarify and solve some of them. Its characteristics are well known:

(i) Power 60 MW(th) (up to 1100 kW/l); (ii) Sodium outlet temperature up to  $580^{\circ}$ C; and (iii) Uranium dioxide fuel.

Building and assembly work is now going on in Melekess, and the reactor is due to come into operation in 1968. The Melekess Atomic Reactor Institute is becoming increasingly involved in work on the use of fast reactors for nuclear power generation.

In our view, the successful introduction of the BN-350 and BOR-60 reactors are the most important objectives for the immediate future in the fast reactor field.

The BR-5 reactor continues to work at its full capacity of 5 MW, using monocarbide fuel. Burn-up in the monocarbide has exceeded 2%. The first core elements with plutonium oxide reached a burn-up of 6-7%. Many test elements are at present also being irradiated. Some of the core elements have developed leaks. Experiments are being carried out to determine that part of the fission product output which differs from the output obtained with oxide fuel. Changes were also observed in the power coefficients. All these problems require further investigation.

The BR-1 reactor was used as neutron source in experiments carried out on the Doppler effect in  $^{238}$ U with sample temperatures up to 2000°K, and the results of these experiments are reported in paper SM-101/63. The reactor is now being re-equipped for carrying out similar Doppler effect measurements, with heating of the medium surrounding the sample.

Measurements of the Doppler effect in the harder spectrum in the BFS core are also under way.

This critical assembly was used to investigate physics problems of reactors with absorbing control rods. Methods were meanwhile developed

<sup>&</sup>lt;sup>1</sup> This presentation and the ensuing discussion took place at the beginning of Session III but is reproduced here because of the nature of the subject matter.

for calculating heterogeneous systems. Two reports presented at this symposium (SM-101/61 and 62) have dealt with methods of calculating the heat release distribution and heterogeneous effects and with a comparison of the calculated and experimental results.

The purpose of recent work on the BFS has been to clarify the reasons for discrepancies between calculated and experimental results on neutron lifetime. In future the BFS will be operated in conjunction with an electron cyclotron in order to carry out various time-of-flight experiments and to measure the spectra of fast reactors of varying composition. Sodium blocks have been developed for measuring the sodium void effect. It should be noted that the nuclear constants, originally accepted on the basis of macro-experiments on a number of elementary characteristics, are showing themselves more and more open to criticism. A new set of constants is being drawn up, incorporating the latest developments in nuclear data. It is furthermore proposed to define the degree of accuracy needed in establishing the group constants, so that the reactor characteristics may be determined to the required accuracy.

There is a constantly growing need for effective and economical methods of reactor calculation in real geometry. A considerable contribution to methodology has been made by the Physics Institute of Moscow University. The generalized method of conditional separation of spatial variables makes it possible to determine the integral characteristics of the neutron field in two-dimensional reactors. On the basis of synthetic methods, multigroup problems can be converted into problems involving few groups. Furthermore, a method for solving the kinetic equation in plane multi-layer geometry has been worked out, making it possible to solve the heterogeneity problem in BFS-type critical assemblies. The development of new economical synthetic methods makes it possible to draw up several comprehensive, optimized programmes.

W.B. LOEWENSTEIN: Could Mr.Krasnoyarov please elaborate on the changes in power coefficient observed during operation of BR-5?

N.V. KRASNOYAROV: The preliminary information to date is as follows: (i) The fast region of the power effect is found to be non-linear; (ii) The reactivity effects observed for the positive and negative power jumps are different.

These phenomena had not previously been noted during investigation of a reactor with fresh monocarbide fuel.

A. FERRARI: Several speakers have made reference to attempts at writing three-dimensional diffusion codes. I wonder whether such an unwieldy code is really necessary, and whether synthesis methods alone would not be sufficient in dealing with the three-dimensional problems which arise in practice.

B.I. SPINRAD (Chairman): Three-dimensional synthesis codes have been successfully used for a variety of problems, but one must be careful to prove the method for each system to which synthesis is applied. Such a method depends on minimizing a variational functional, but attempts to find a theoretical functional which minimizes variations in flux shape have met with no success. A reactivity criterion is therefore usually employed, and it is necessary to verify for each reactor that the minimization of reactivity error gives a proper flux shape. This is done by comparing the results of synthesis calculations with a number of critical experiments, or exact three-dimensional calculations, on systems of sufficient geometrical complexity to cover the required range.

F. STORRER: We used an energy synthesis code to deal with the interface between two core regions with similar spectra, as reported to the Argonne National Laboratory (ANL) Conference of October 1966, and the reactivity we found was in perfect agreement with that calculated by an alternative method both more exact and more costly in computer time. I should be interested to hear which synthesis codes — be they space or energy methods — are being studied at present in the USSR.

M.F. TROYANOV: Synthetic methods and various applications thereof are currently under study in the Engineering and Physics Institute of the USSR. A neutron-field synthesis in two-dimensional r, z geometry has been worked out, enabling the integral characteristics and neutron balance in a multi-zone reactor to be reliably predicted. This method of calculation saves substantially on the time needed for the accurate twodimensional calculation, without adversely affecting the determination of the integral characteristics. A neutron field synthesis in two-dimensional x,  $\mu$  geometry (a solution of the kinetic equation) has also been carried out; we used this variant of the method in calculating heterogeneous effects in BFS-type critical assemblies. Finally, methods of spaceenergy synthesis have been developed for dealing with two-dimensional problems with a small number of groups. These methods have been published in the collected papers of the Engineering and Physics Institute for 1965 and 1966.

## NUCLEAR DATA FOR FAST REACTORS (Session II and Session III, Part 1)

Chairmen: S. YIFTAH and K.E.J. WIRTZ

## NEUTRON CAPTURE AND FISSION CROSS-SECTION DATA IN THE keV ENERGY RANGE Some new measurements and renormalizations\*

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#### Abstract

NEUTRON CAPTURE AND FISSION CROSS-SECTION DATA IN THE keV ENERGY RANGE: SOME NEW MEASUREMENTS AND RENORMALIZATIONS. Some new measurements of capture cross-section have been performed using the 3-MeV Karlsruhe pulsed Van de Graaff accelerator. A major effort was undertaken to measure the absolute gold capture cross-section as a function of energy between 25 and 500 keV in order to establish a reliable standard for capture and fission cross-section renormalizations. To do this, a well-collimated pulsed beam of monoenergetic neutrons produced in a thin Lithium target at 80° to the proton beam direction was used to irradiate a 0.1-cm thick sample in the centre of a large liquid scintillator which detects the prompt capture y-rays. The relative neutron flux was measured with a 'grey neutron detector' which is relatively insensitive to neutron energy variations in the energy range of these measurements. The gold cross-section shape was normalized at 30 keV to an absolute value fitted from various recent experiments.

By a similar technique, the capture cross-section of <sup>238</sup>U in the energy range 25 - 500 keV was determined. Furthermore, cross-sections of several medium and heavy weight nuclei which may be of some interest to reactor design (Nb, Mo, Cs, Hf, Ta, W, Re) were determined by a time-of-flight method, using our gold value as a standard. The data are presented and some implications of the new data on reactor calculations are reviewed.

#### 1. INTRODUCTION

Radiative neutron capture is one of the most important nuclear processes relevant to the neutron balance in fast reactors. There has thus been a great effort in many laboratories to measure capture crosssections, particularly in the keV energy range. Nevertheless, the situation is still far from satisfactory. The main regions where data are incomplete, as discussed recently within international data committees, are the following:

(a) Although cross-section ratios can be measured with sufficient accuracy in many cases, the deduction of absolute cross-section values has in most cases not been carried out consistently owing to the lack of a proper and internationally recognized standard cross-section.<sup>1</sup> This holds particularly in the energy range 10 - 100 keV where flux measurements (and thus standard cross-section determinations) are difficult to carry out.

<sup>\*</sup> Work performed within the association in the field of fast reactors between the European Atomic Energy Community and Gesellschaft für Kernforschung m.b.H., Karlsruhe.

<sup>&</sup>lt;sup>1</sup> This is also true for fission cross-sections.

(b) For some of the most important reactor materials, especially the fertile and fissile nuclei, a very high accuracy – in some cases down to  $\pm 1\%$  – is required. Even if standard cross-sections were known to this accuracy, these requests could not always be fulfilled since the cross-section ratio measurement methods are not that accurate.

(c) Cross-section ratio measurements on very small samples or on highly radioactive materials are difficult to perform and require the development of special techniques. Similarly, accurate measurements on nuclei with small capture cross-sections, e.g. lighter structural materials like Fe, cannot be carried out easily.

(d) There is finally a number of nuclei for which adequate measuring methods as well as samples are available but which hitherto have just escaped attention. To these belong many stable fission products and control materials.

It is obvious that these problems, whose solution might require a total effort of several hundred man-years, can only be tackled by close international co-operation between many laboratories. Some first steps in this direction have recently been taken by the European-American Nuclear Data Committee.

To contribute to some of the above problems, a programme has been launched at the Karlsruhe 3-MeV pulsed Van-de-Graaff accelerator which will be briefly reviewed in this paper. So far, a large part of our effort has been spent on the standardization problem where we have remeasured the absolute capture cross-section of gold between 25 and 500 keV to an accuracy of about  $\pm$  5%. We propose to use this cross-section curve as a standard for further renormalizations of capture and fission cross-sections. The results of this experiment are described in section 2. Our method for determining the gold cross-section is rather tedious and time-consuming, yet we believe that some cross-sections like <sup>238</sup>U capture and <sup>235</sup>U fission are so fundamental to fast reactor calculations that they warrant a direct determination by essentially the same method to avoid additional errors due to a cross-section ratio measurement. As a first step along these lines, we have redetermined the <sup>238</sup>U capture cross-section in the energy range 25 - 500 keV (section 3). The standard time-of-flight technique using a large liquid scintillator tank for capture cross-section measurements has also been further developed in our laboratory; some results on heavy structural materials are given in section 4.

In conclusion, section 5 contains some of the implications of these new data for reactor calculations and some general comments on future capture cross-section work.

#### 2. ABSOLUTE CAPTURE CROSS-SECTION OF GOLD

This experiment was performed in two steps. First, the shape of the cross-section curve between 25 and 500 keV was measured by a new technique. Then, this relative cross-section curve was normalized at 30-keV neutron energy, using results from several independent absolute measurements.

The experimental set-up for the shape measurement is shown in Fig. 1. A thin gold foil was irradiated by a collimated beam of nearly monoenergetic neutrons, obtained at an angle of 80° from the  $^{7}$ Li(p, n)<sup>7</sup>Be reaction (target thickness: 12-18 keV for low energies, 12-40 keV in the middle energy region and 40-100 keV for high energies). The gold sample was located within a 1.1-m-diam. large liquid scintillator tank [1,2] which was used to determine the relative capture rate. The pulsedbeam - time-of-flight method was used for background discrimination. The neutrons transmitted through the gold foil were thermalized and totally absorbed in a 'grey neutron detector' [3]. This is a paraffin pile, size  $60 \times 60 \times 60$  cm, with a  $10 \times 10 \times 30$  cm beam entrance hole, located at the end of the neutron path. Capture of thermalized neutrons by hydrogen leads to the well-known 2.2-MeV  $\gamma$ -ray; since  $\gamma$ -ray absorption in paraffin at this energy is small, the photopeak counting rate of a NaI (T1) detector located near the pile is very nearly proportional to the capture rate and thus to the impinging neutron current. This detector has to a first approximation an efficiency curve which is independent of neutron energy; to a higher approximation small deviations from the flat efficiency curve exist which can, however, be calculated [3].



FIG.1. Experimental arrangement for cross-section shape determination.

The relative capture cross-section follows directly from the counting rate of the large liquid scintillator tank and from the corrected counting rate of the grey detector. However, some slight corrections have to be applied in addition: relation between average cross-section and cross-section at an average energy; resonance self-shielding and multiple scattering within the gold foil; efficiency change of the large liquid scintillator whose bias was set at about 3.6-MeV photon energy with neutron energy; air scattering of neutrons between the gold foil and the grey detector; appearance of the second neutron group from the <sup>7</sup>Li(p, n) <sup>7</sup>Be reaction above  $\sim$  380-keV neutron energy. These corrections, together with a detailed discussion of the experimental approach, will be published elsewhere [4].

The absolute normalization of the relative cross-section curve was performed at 30-keV neutron energy. Table I lists the results of several

Ме	thod		Average value (mb)	
Flux det.	Capture rate det.	Reference		
Associated activity	Activity	[8,18]	598 ± 30	
Integral (MnSO4-bath)	Activity	[18 - 20]	596 ± 20	
Relative ( <sup>235</sup> U-fission)	Activity	[21]	608 ± 40	
Relative ( $^{10}$ B-n, $\alpha$ )	Prompt capture γ-rays	[22 - 24]	587 ± 21	
Shell transmission		[25 - 28]	614 ± 37	

TABLE I. GOLD CAPTURE CROSS-SECTION VALUES AT 30 keV

more recent determinations at this energy from five independent methods. The table includes data taken with Sb-Be sources which were transformed to 30 keV assuming  $E_n = 22.8$  keV [5] for Sb-Be and a cross-section ratio of 0.915 ± 0.040 [2]. The fitted best value from all these results is

 $\sigma_{n} (Au) (E_n = 30 \text{ keV}) = 0.596 \pm 0.012 \text{ barn.}$ 

The gold capture cross-section as a function of energy, normalized with the above value, is shown in Fig.2.



FIG.2. Gold capture cross-section as a function of neutron energy.

We have carried out a further independent determination of the gold capture cross-section shape in the energy range 10 - 150 keV. A timeof-flight method with neutrons from a thick <sup>7</sup>Li target was used. The capture rate in a gold foil was measured with the large liquid scintillator tank, while the relative neutron flux was simultaneously determined with

a thin boron-10 slab viewed by NaI (T1) scintillators or, in a different run, by a <sup>6</sup>Li glass detector. Both detectors were placed at the exit of the large liquid scintillator tank. The boron-10 n,  $\alpha\gamma$  cross-section as recommended by Spaepen [6] and the <sup>6</sup>Li n.  $\alpha$  cross-section from the Breit-Wigner fit of Bergström et al. [7] were used for the calculation of the neutron detector efficiencies. In these calculations, corrections for multiple scattering of neutrons within the detector were applied. The measurement yielded however only the relative shape of the gold crosssection since no effort was made to determine the absolute efficiency of the neutron detectors. The shape curves were again normalized to a value of 596 millibarns at 30 keV. The resulting curves are compared to the grey detector results in Fig. 3. Note that the values found relative to boron and to lithium agree very well among themselves. Since they were taken with higher resolution than the grey detector data they show some structure. On the average, they agree very well with the grey detector data, at least below 80 keV. The deviations above 80 keV are small and well within the limits of experimental error.



FIG. 3. Comparison of gold cross-section shape as measured with grey detector (-----), <sup>6</sup>Li glass scintillator (O) and <sup>10</sup> B slab ( $\Delta$ ).

A detailed comparison of our new standard cross-section as shown in Fig.2 with the results of previous experiments is given in Ref. [4]. Briefly speaking, there is good agreement of the shape with most of the recent determinations by various methods. There is also good agreement in the absolute values over the whole energy range with a recent experiment by Harris et al. [8] which was performed by an absolute method and is, like ours, independent of reference cross-sections. On the other hand, there is a striking disagreement in the absolute cross-section values above 100 keV with several other experiments which are based essentially on a measurement of the ratio  $\sigma_{n,\gamma}^{Au}/\sigma_{n,f}^{zs}$ . This holds in particular for the recent experiments of Barry [9] and of Vaughn et al. [10] which are in themselves consistent but yield, when evaluated with White's [11] <sup>235</sup>U fission cross-section, a gold capture cross-section which is always about 15% higher than our results.

#### 3. <sup>238</sup>U CAPTURE CROSS-SECTION

The  $^{238}$ U n,  $\gamma$  cross-section could be determined by measuring the value relative to gold and subsequent normalization, using the data of Fig.2. Since this would introduce several errors, i.e. the errors in the



FIG.5. Comparison of experimental values of <sup>238</sup>U capture cross-section with compiled values.

standard cross-section and in the ratio measurement would combine, we have determined it by the same method as described for gold in the previous section. The normalization of the shape curve which had been found by the grey detector - large scintillator tank method was again done at 30 keV. Therefore, an absolute cross-section measurement was performed at the <sup>7</sup>Li(p, n) <sup>7</sup>Be threshold [12], using an activation method and a neutron flux determination by the associated <sup>7</sup>Be activity method.

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The induced  $^{239}\mathrm{Np}$  activity (T $_{\frac{1}{2}}$  - 2.346 d) was determined by two independent methods ( $\gamma$ -ray counting with a 30 cm<sup>3</sup> Ge(Li) detector; 106-keV  $\gamma$ -X-ray coincidence counting method), the detectors being calibrated by using an absolutely calibrated  $^{243}\mathrm{Am}$  source. The result at 30 keV (i.e. for a beam with an average energy of 30 keV and a distribution with a half-width of 15 keV) is  $\sigma_{n,\gamma}^{238U}$  (En = 30 keV) = 0.479  $\pm$  0.014 barn. The  $^{238}\mathrm{U}$  capture cross-section as normalized with this value is shown in Fig.4. The error is about 5% at the lower energy limit and increases to 9% at the upper end.

In Fig. 5 we compare our results with evaluated data as taken from recent compilations of Parker [13], Schmidt [14] and Stehn [15]. The agreement with Stehn's data below 100 keV is quite satisfactory; our data are distinctly lower than those of all three compilers in the energy region above 100 keV. The implication of this deviation for some reactor calculations will be discussed in section 5.

#### 4. SOME RENORMALIZATIONS OF CAPTURE CROSS-SECTION DATA

Using a time-of-flight method and the large liquid scintillator tank, we measured the capture cross-section, relative to gold, for a number of medium-weight and heavy nuclides in the energy range 10 keV - 150 keV. Preliminary data for Ta, W, Mo, In, Re, Hf, Cd, Ag, Pd, Nb and Cs were presented at the 1966 IAEA nuclear data conference [2]. These data have now been re-evaluated, using the new gold capture cross-section data.<sup>2</sup> Re-evaluated data for those nuclei which may be of some interest for reactor calculations are shown in Fig.6. The data are corrected for resonance self-shielding and multiple scattering. Their accuracy (including the standard cross-section error) is about 10% over the whole energy range.

Preparations for further measurements of relative cross-sections, especially for stable fission products and for lighter structural materials like iron are being made at our laboratory.

## 5. SOME IMPLICATIONS OF THE NEW DATA FOR FAST REACTOR CALCULATIONS.

To check the influence of the new <sup>238</sup>U capture cross-section on reactor parameters, the new data were converted into group constants and included on a trial basis into the KFK-SNEAK-26-group cross-section set [16]. This set is based on Schmidt's cross-section curves [14]. As can be seen from Fig. 5, the Schmidt data are distinctly higher than ours above 100 keV; there is a corresponding decrease of the group crosssections in this energy range.

The modified set was used for calculations of the SNEAK 3A-1 critical assembly [17], a 670 litre steam-cooled  $^{235}U/^{238}U$  system. Whereas the

<sup>&</sup>lt;sup>2</sup> Strictly speaking, the measured cross-section ratios were evaluated using a gold cross-section curve intermediate between the two sets of curves in Fig.3; the difference between this and the grey detector result of Fig.2 is, however, unimportant.



FIG.6. Capture cross-sections for Nb, Hf, Mo, Ta, Cs, W and Re.

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unmodified KFK-SNEAK-set slightly underestimates reactivity ( $k_{eff}$  = 0.997 for a system of critical size), the modified data increase the multiplication factor by about 1%. At the same time, they lead to a reduction of the conversion ratio by about 5%. It is possible that the increase in reactivity would be partly compensated if the <sup>235</sup>U fission cross-section were reduced by normalizing measured gold capture to <sup>235</sup>U fission cross-section ratios by using our new gold cross-section standard. Such investigations are at present under way but it seems premature to draw any conclusions.

In any case, we feel that it is highly desirable to discuss the problems of keV capture and fission cross-section normalizations in close connection with criticality calculations. In particular, interpretations of measurements on pulsed subcritical systems where the ratio of absorption to leakage can be arbitrarily shifted, may be helpful.

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#### DISCUSSION

W. HART: Peter White's fission cross-section measurements for  $^{235}$ U in the 100-keV energy region are quoted as being accurate to about 2 or 3%. What is the corresponding accuracy associated with the  $^{235}$ U fission cross-sections obtained from your measured gold capture cross-sections and reported  $\sigma_f^{235}$ // $\sigma_c^{Au}$  ratios?

K.H. BECKURTS: The accuracy of our gold measurements in that energy range is about 4-5%. However, if they are compared indirectly, in the manner you indicate, with White's values, the discrepancy between the two is 12%, which is definitely outside the limits of mutual error.

J.L. ROWLANDS: I note that you find that reduction of the  $^{238}$ U capture cross-section results in a 5% reduction in the conversion ratio. We find that a reduction in the  $^{238}$ U capture cross-section causing a 1% reduction in reactivity changes the breeding ratio by less than 1%, in fact.

K.H. BECKURTS: I shall refer this question to Dr. Küsters, who carried out these calculations. I might add, however, that after we changed the <sup>235</sup>U fission and capture data using our gold renormalization, the total change in conversion ratio was very small.

H. W. KÜSTERS: Our method of handling the replacement of the low Pönitz-Menlove <sup>238</sup>U capture data was to introduce them into the SNEAK set, and then to perform a fundamental mode calculation, achieving criticality by buckling iteration. In this case there is definitely reduction of the conversion ratio. Dr. Rowlands is, I think, referring to the case where criticality is achieved by altering the enrichment. The change in conversion ratio is then certainly negligibly small.

T.A. PITTERLE: In the reported calculated  $^{239}Pu/^{235}U$  and  $^{238}U/^{235}U$  fission ratios, were the  $^{239}Pu$  and  $^{238}U$  fission cross-sections used renormalized to your new  $^{235}U$  cross-sections?

K.H. BECKURTS: No, they were not.

T.A. PITTERLE: Most of the reported change in these fission ratios would then be due solely to lowering of the <sup>235</sup>U fission cross-section and would not really indicate an improvement in the calculated spectrum obtained with the KFK-SNEAK set.

Consistent use of  $^{239}$ Pu/ $^{235}$ U and  $^{238}$ U/ $^{235}$ U fission ratios would probably reduce the difference between the calculated fussion ratios of the cross-section sets.

K.H. BECKURTS: Quite likely so, but one should note that on our results the agreement for  $\sigma_c^{28} U/\sigma_f^{26} U$  is also improved and that in this case both cross-sections were re-adjusted.

## ENERGY- AND TEMPERATURE-DEPENDENT CAPTURE MEASUREMENTS BELOW 30 keV SUPPORTING DOPPLER EFFECT CALCULATIONS\*

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#### Abstract

ENERGY - AND TEMPERATURE-DEPENDENT CAPTURE MEASUREMENTS BELOW 30 keV SUPPORTING DOPPLER EFFECT CALCULATIONS. Energy- and temperature-dependent capture measurements below 30-keV neutron energy were performed in natural uranium, tungsten, and tantalum using the slowing-down time spectrometer technique. The experimental set-up used for the experiments consists of a lead block of 1.3 m side length containing two experimental channels of 10×10 cm<sup>2</sup> cross-section. Into the first channel the target of a 14-MeV neutron generator is introduced, whereas the second channel is used for insertion of the heated samples. Pulses of 14-MeV neutrons, having a pulse width of about 1 µs, are used. The neutron energy is degraded first by inelastic collisions; afterwards only elastic collisions take place so that a specific relationship holds between mean neutron energy in the lead pile and the time after occurrence of the neutron pulse. Because of this time-energy relation a time analysis procedure for the detector counts is applied. Because the energy range below 30-keV neutron energy is most interesting for Doppler-effect investigations the slowing-down time spectrometer is used to measure the capture ratios of hot-to-cold samples of natural uranium, tungsten, and tantalum. Thin samples were heated to different temperatures for this purpose, and the capture y-rays were detected by proportional counters. Because hot-to-cold capture ratios are measured a knowledge of the neutron flux is not necessary; therefore, a direct comparison of calculated and measured temperature-dependent cross-sections is possible. A theoretical analysis of the experimental data for uranium is given.

#### 1. INTRODUCTION

The idea of measuring temperature- and energy-dependent reaction rates was considered necessary to support the analysis of integral Doppler experiments performed in fast critical assemblies. This support concerns only the neutron and nuclear physics aspects of Doppler-effect analysis. The additional reactor physics complications are avoided and thus the problems for a detailed comparison of experiment with theory are reduced. Besides the Doppler-effect of fissile and fertile reactor materials, the study of high-temperature metals like tungsten and tantalum is also of interest in advanced high-temperature reactors.

The aim of this experiment was the measurement of energy-dependent reaction rates below 30 keV at different sample temperatures. For this purpose the neutron slowing-down time spectrometer [1, 2] was used. This operates as follows: fast monoenergetic neutron bursts of short

<sup>\*</sup> Work performed within the association in the field of fast reactors between the European Atomic Energy Community and Gesellschaft für Kernforschung m.b.H., Karlsruhe

duration are slowed down in a material of high mass number in which they maintain a relatively narrow energy distribution. Therefore, a relation holds between slowing-down time and average neutron energy. The moderator, a lead cube, contains an experimental channel into which the sample under investigation is positioned. Neutron captures in the sample result in spontaneous  $\gamma$ -rays from the decaying nuclei. These capture  $\gamma$ 's are measured time-dependent by proportional counters. By this procedure hot-to-cold reaction rate ratios between 0.2 and 30 keV were measured in similar time-dependent neutron spectra. Because of this the neutron flux cancels almost completely in the hot-to-cold ratio formed, which is an important advantage of this experiment. Apart from this the resonance energy region of interest for Doppler experiments coincides with the energy range covered by the slowing-down time spectrometer, so obtaining an optimum energy-dependent signal of the Doppler-effect at different temperatures.

This type of experiment is well suited to check theoretical analysis regarding uncertainties in nuclear data and to correct calculation of temperature-dependent cross-sections in the statistical region of resonances. This treatment includes resonance interference and overlap effects of different isotopes present in one mixture. The results from these experiments are a helpful tool for the analysis of energy-integrated foil activation measurements or other Doppler experiments in fast reactors.

#### 2. EXPERIMENTS AND RESULTS

#### 2.1. Description of experimental set-up

A sketch of the experimental set-up is shown in Fig.1. The main components are the neutron generator and the lead cube of 1.3 m side length. Highly purified lead (99.99%) was used for the slowing-down time spectrometer. Two channels of  $10 \times 10 \text{ cm}^2$  cross-section were cut out of the lead cube, one for the target and one for the sample under investigation. The lead cube was surrounded by a 1-mm-thick Cd sheet to prevent re-entrance of slow neutrons. Short neutron bursts were generated by the reaction T(d, n) $\alpha$  with a pulsed Cockroft-Walton type accelerator. The titanium-tritium target placed in the centre of the lead cube was bombarded by the deuterons accelerated up to 150 keV. The ions from a Penning-type source were accelerated in d.c. operation and the ion pulses of 0.4 to 1.5  $\mu$ s duration were formed by deflection plates as indicated in Fig.1. The pulse repetition rate was 2 kc/s for nearly all experiments.

#### 2.2. Detectors, furnaces and electronics

Three experimental runs, two at different sample temperatures and one background measurement, are necessary to evaluate the hot-to-cold capture ratio. To make intercomparison between the three runs the neutron source strength had been monitored by a  $BF_3$  proportional counter shielded with Cd against slow neutrons. Its position is indicated in Fig.1.

Figure 2 shows the cross-sections of the two different furnaces used in these experiments. In assembly A the sample is surrounded by the detector in contrast to assembly B. The detectors were proportional counters of 30 cm sensitive length and 10 mm diam. at 1 mm wall thickness. They were filled with argon at a pressure of 760 mmHg. Two assemblies A and B were constructed because different aspects had to be taken into account. One aspect is the optimum detector efficiency obtained by maximum solid angle in order to have reasonable counting times. Another aspect concerns a maximum signal-to-background ratio. which is of utmost importance if the sample cross-section is low. Assembly A is best suited for large cross-section samples because it has approximately  $4\pi$  geometry. Assembly B, on the other hand, offers maximum signal-to-background ratio. The disadvantage, however, is the reduced detector efficiency. For the geometries shown in Fig.2 the background is reduced by a factor of 10 from assembly A to B. The corresponding efficiency (solid angle) is only reduced by a factor of about 4 from A to B. This has been found from calculations. Therefore, assembly B had to be used to measure capture in <sup>238</sup>U.



FIG. 1. Set-up of neutron slowing-down time spectrometer.

For the furnace constructions in Fig. 2 some additional information is given: all furnace material including the electrical feed-throughs and proportional counter tubes were fabricated from AlMg3, an alloy (97% Al, 3% Mg) having an extremely low capture cross-section. The volume between the inner and outer furnace wall was evacuated up to 10<sup>-4</sup> Torr by a diffusion pump to avoid heat convection. The sample is heated by 1-mm Thermocoax wires. The heating power necessary to achieve a sample temperature of 700°K was 12 W for assembly A and 70 W for assembly B. Temperature was measured by NiCr-Ni thermocouples. All inner surfaces were polished in order to have high reflection of heat radiation.



FIG.2B. Profile of furnace assembly B.

The electronic circuitry is shown in Fig.3. The monitor channel is conventional electronics. The special kind of signal channel will be explained in more detail. Actually two detectors feed one linear channel with pulses. The signals of the five channels are mixed and afterwards time-analysed. These five channels were found to be necessary because during and shortly after the neutron bursts high  $\gamma$ -flux impinges onto the detectors. Using five channels instead of one results in a considerable reduction of pulse pile-up. Conventional non-overload amplifiers have – for these high-amplitude input pulses – long recovery times and base line shifts. The mixer of the five-channel circuitry delivered fast output signals of 100-ns pulse width and had a time resolution of the same order. The pulses were finally analysed by a fast shift register<sup>1</sup> [3]. The maximum counting speed together with the multiscaler storage system was  $5 \times 10^6$  pulses/s in every channel. Because of deadtimes of the order of 10  $\mu$ s, conventional time analysers could not be used for these measurements as they cannot handle the counting rates of about  $2 \times 10^5$  pulses/s during and shortly after the neutron bursts. In this set-up the limiting factors concerning deadtime were the amplifiers and discriminators used in the counting channels having about 2 to 3  $\mu$ s deadtime, so in this respect the set-up was not optimum.

In addition to the need for extremely short deadtimes, the total drift in count rate should be better than 0.5% per 10 hours, since measuring times of several hours are possible only in this way. Long measuring times, however, are necessary to achieve good statistics. The main instabilities result from drifts of discriminator levels and less from gain variations of amplifiers. In Fig.4 the gamma pulse height spectrum of a proportional counter is shown. The discriminator level has been set according to the indication in Fig.4. The necessary long time behaviour was even better than 0.5% in our arrangement.

#### 2.3. Energy calibration of slowing-down time spectrometer

The relation between average neutron energy and slowing-down time for an elastic scattering medium is given by:

$$\overline{E}(t) = \frac{m_N A^2 \lambda_s^2}{2} \frac{1}{(t + \frac{A \lambda_s}{V_s})^2}$$
(2.1)

or

 $= \sqrt{\frac{C}{E}} - t_o$ 

(2.2)

with

The symbols are explained in the nomenclature given at the end of the paper. To calibrate the spectrometer in the energy range under investigation two different materials with resonances of known energies were measured. A plot of slowing-down time versus  $1/\sqrt{E}$ , shown in Fig.5, is best suited to compare the constant C with earlier calibrations [2]. The result is C = 185 keV  $\mu$ s<sup>2</sup> compared to 179 keV  $\mu$ s<sup>2</sup> in Ref.[2]. The resonance energies were taken from BNL-325.

 $= \frac{m_N A^2 \lambda_s^2}{2}$ 

 $= \frac{A \lambda_s}{v}$ 

<sup>1</sup> Manufactured by BORER and Co., Solothurn, Switzerland.

t







FIG.4. Gamma spectrum of a proportional counter.

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To avoid the influence of higher harmonics of the spatial neutron distribution on energy calibration the  $\gamma$ -detectors were positioned at zero crossing of the third harmonic as indicated in Fig.1.



FIG.5. Dependence of slowing-down time t on  $1/\sqrt{E}$ .

#### 2.4. Resolution of slowing-down time spectrometer

The energy resolution is determined on the one hand by the slowingdown phenomena, on the other hand by uncertainties in the time measurement and the extended neutron pulse width. The following three slowingdown phases are mainly responsible for the energy resolution of the spectrometer:

- (1) Inelastic scattering above 0.5 MeV
- (2) Asymptotic energy distribution resulting from the statistics of scattering below the first excited level
- (3) Thermal motion of lead atoms at eV energies.

The calculated energy resolution as function of slowing-down time is plotted in Fig.6, taking into account all effects mentioned above (neutron pulse width  $1.5 \ \mu$ s).

The resolution of the spectrometer is appropriate for measuring the energy-dependent resonance integral in the statistical energy region from 0.2 to 30 keV. As is more fully explained in section 3 the following condition must hold:

$$\overline{\Gamma_n^{s}}$$
,  $\Gamma_\gamma \ll \overline{D^{s}} \ll \Delta E \ll E$ 

(2.3)

It can be seen from Fig. 6 that condition (2.3) is satisfied.



FIG.6. Slowing-down time spectrometer resolution curve.

#### 2.5. Deadtime effects and test measurements

The high counting rates during and shortly after the neutron bursts (the reasons for this effect are summarized in Ref. [2]) could not be registered without counting losses owing to the deadtime limit explained in section 2.2. To investigate the influence of deadtime effects on the hot-to-cold  $\gamma$ -ray ratio the following experiment was carried out. The basic idea of this experiment is to simulate the increased counting rate from the Doppler-effect by higher neutron intensity. The procedure is explained by the following formula:

$$\frac{S_{I_1}(t) - B_{I_1}(t)}{S_{I_2}(t) - B_{I_2}(t)}$$
(2.4)

where S is the signal plus background at  $I_1$  and t, B the background at  $I_1$  and t,  $I_i$  the neutron intensity i, and t the slowing-down time. This ratio is very similar to the hot-to-cold  $\gamma$ -ray ratio of a real experiment defined by:

$$R = \frac{H(t) - B(t)}{C(t) - B(t)}$$
(2.5)

where H is the signal plus background of hot sample at t, C the signal plus background of cold sample at t, and B the background at t.

The only difference between Eq. (2.4) and Eq. (2.5) is that the background depends on neutron intensity in Eq. (2.4). However, Eqs (2.4) and (2.5) are almost equal in our case and a measurement approximates the deadtime effect closely. In Fig.7 the result of a deadtime experiment is shown in which tantalum has been measured at two neutron intensities differing by 9.5%. Up to the fifth channel the deadtime loss is large and affects the accuracy of hot-to-cold  $\gamma$ -ray ratios appreciably. Consequently we reduced the deadtime effects in the first five channels by running the neutron generator at reduced power, which means 110 kV accelerating voltage and 2 mA target ion current for nearly all measurements.



FIG. 7. Ratio of gamma rays at different neutron intensities.



FIG. 8. Gamma ratio of a shielding experiment.

Another effect which was investigated concerns the  $\gamma$ -shielding of the detectors by the samples to be measured. Very thick samples absorb  $\gamma$ -rays from the lead and, therefore, change background. Figure 8 shows the result of such a shielding experiment where the sample was replaced by a 0.2-mm-thick lead foil. The thickness of this foil was sufficient to simulate the  $\gamma$ -absorption effect of the samples investigated. The measurement shows that the absorption effect on background can be neglected. Finally, it was necessary to measure the ratio of heated-to-cold empty furnace to make sure that the heating itself does not contribute to the hot-to-cold ratio of a sample measurement. For a temperature change of 400 degC the result is plotted in Fig.9. This includes the influence of furnace dislocations by heating. This, too, does not affect the results.



FIG. 9. Hot-to-cold background y-ray ratio versus slowing-down time.

#### 2.6. Measurements with <sup>238</sup>U

For <sup>238</sup>U the ratio of the  $\gamma$ -counting rates obtained with the heated sample to the  $\gamma$ -counting rate with the sample at room temperature, after subtraction of background(hot-to-cold  $\gamma$ -ray ratio), is shown as a function of average energy or slowing-down time in Figs 10 and 11, respectively. The experimental conditions – also used for the theoretical calculations according to section 3 – are given in Table I. The experimental results are shown by crosspoints and the theoretical calculation is presented by the dashed curve. The error margins given are based only on statistics at 68% confidence level. They are evaluated from the formula

$$\frac{\Delta R}{R} = \frac{1}{(H-B)(C-B)} \sqrt{(C-B)^2 H + (H-C)^2 B + (H-B^2)C}$$
(2.6)

which is derived from Eq. (2.5) with the error propagation law. To reduce the error margins of the experiment, a faster electronic circuitry for the linear channels is necessary and now under construction for future experiments. A statistical error in R-1 of about 5 to 10% is aimed for. In spite of the limited accuracy the experimental data seem to indicate the possibility of interpreting the capture rate increase (see section 3) by statistical theory down to several hundred eV.

 $^{238}$ U is of special interest in Doppler investigations in fast reactors and justifies further efforts to improve accuracy. In this context, however, a number of difficulties have to be mentioned which are particular to  $^{238}$ U. The main difficulty arises from the low signal-to-background ratio, which means that gammas from lead contribute to the counting rate almost as much as the capture gammas of the sample itself. The



FIG. 10. Hot-to-cold <sup>238</sup>U capture  $\gamma$ -ray ratio versus  $\overline{E}$ .



FIG. 11. Hot-to-cold <sup>238</sup>U capture  $\gamma$ -ray ratio versus t.

disadvantage from this effect follows directly from the first term of Eq.(2.6). To minimize the background the  $\gamma$ -rays resulting from the decays of <sup>238</sup>U and impurities had been almost eliminated by shielding the detectors with 300- $\mu$ m lead as indicated in Fig.2B. Another difficulty

## TABLE I. PARAMETERS FOR THE <sup>238</sup>U-MEASUREMENT

Hot sample temperature (°K)	750
Cold sample temperature (°K)	300
Foil thickness (µm)	155
Foil length (cm)	25
Foil weight (g)	51.33
σ <sub>p</sub> (barns)	· <b>1</b> 4



FIG. 12. Hot-to-cold tantalum capture  $\gamma$ -ray ratio versus  $\overline{E}$ .

arises from the oxidization of uranium metal at high temperatures. To avoid this we put the foil between two closely spaced aluminium tubes welded at the ends. Because of the small volume left almost no oxidization took place.

#### 2.7. Measurements with tantalum and tungsten

For natural tantalum and tungsten, measured hot-to-cold  $\gamma$ -ray ratios versus energy are shown in Figs 12 and 13. The experimental conditions are shown in Table II. Furnace type A was used for these experiments. The solid lines are fits to the experimental data. Theoretical calculations for both materials have not been done so far. Figure 12 is the average of three runs under the same conditions which had been performed to check reproducibility. The error margins for tantalum are much smaller than for <sup>238</sup>U owing to a better signal-to-background ratio of about 3. The accuracy obtained for tantalum shows the capability of this method for Doppler-effect investigations if the conditions are optimized.



FIG. 13. Hot-to-cold tungsten capture  $\gamma$ -ray ratio versus  $\overline{E}$ .

# TABLE II. PARAMETERS FOR THE TANTALUM AND TUNGSTEN MEASUREMENTS

	Tantalum	Tungsten
Hot sample temperature (°K)	700	700
Cold sample temperature (°K)	300	3.00
Foil thickness (µm)	150	350
Foil length (cm)	30	25
Foil weight (g)	35.19	68.75
· .		

## 3. CALCULATION OF CAPTURE RESONANCE INTEGRALS AT DIFFERENT ENERGIES

### 3.1. The analytic model of the heterogeneous resonance integral

We used the following experimental arrangement. The absorber, an isotope of high mass number, is surrounded totally by the slowing-down material lead. Because the energy width of an absorber resonance is small compared with the energy loss of neutrons by elastic collisions in both the absorber and surrounding moderator, heterogeneous resonance integral theory in narrow resonance approximation can be applied for this situation. We follow the derivation of Dresner [4] for the heterogeneous resonance integral. It is defined in our case as the energyintegrated absorption cross-section which, when multiplied by the flux  $\phi$ that would exist in the absence of the resonance, gives the true absorption rate:

$$N I \not O V = \int_{\Delta E} N \sigma_a \varphi V dE \qquad (3.1)$$

with

$$I = \frac{1}{\Delta E} \int_{\Delta E} \frac{\Sigma_{\mathbf{p}} \sigma_{\mathbf{a}}}{\Sigma_{\mathbf{t}}} dE + \frac{1}{\Delta E} \int_{\Delta E} \frac{(\Sigma_{\mathbf{t}} - \Sigma_{\mathbf{p}}) \sigma_{\mathbf{a}}}{\Sigma_{\mathbf{t}}} P_{\mathbf{o}}(\Sigma_{\mathbf{t}}) dE \qquad (3.2)$$

Equation (3.2) can be rewritten with Wigner's rational approximation for  $P_o(\Sigma_t)$ 

$$I = \frac{1}{\Delta E} \int \sigma_{a}(T) \frac{S + \Sigma_{p}}{S + \Sigma_{t}(T)} dE \qquad (3.3)$$

with  $S = \frac{1}{\overline{\ell}}$ .

This formula used for our experiments is identical to the self-absorption term of the theoretical treatment of Doppler foil activation measurements as performed by Reynolds [5].

## 3.2. Calculation of the <sup>238</sup>U-resonance integral from nuclear data

In this section the method of calculating Eq. (3.3) is specified. The range of calculation has been limited to the statistical energy region because this mainly contributes to the Doppler effect in fast reactors. To show the relationship with the <sup>238</sup>U foil activation experiments which were also carried out at the STARK-reactor [6], we have restricted ourselves to capture processes in <sup>238</sup>U. Capture in monoisotopic nuclides like <sup>181</sup>Ta are treated similarly.

For <sup>238</sup>U the three resonance series are as follows:

1	0	1	1	
j	1/2	1/2	3/2	
s	1	2	3	

The resonances are described in the multi-channel one-level approximation by:

$$\sigma_{t}(x) = \sigma_{oc} \left( \frac{1}{1+x^{2}} \cos 2\delta_{1} + \frac{x}{1+x^{2}} \sin 2\delta_{1} \right) + \sigma_{p} \qquad (3.4)$$

$$\sigma_{\mathbf{a}}(\mathbf{x}) = \sigma_{\mathbf{0}\mathbf{a}} \frac{1}{1+\mathbf{x}^2}$$
(3.5)

with

At low energies the asymmetric term in Eq.(3.4) disappears because of small  $\delta_1$  and will be neglected in the following treatment.

Conventional methods [4] have been used to calculate the temperature dependent Doppler-effect, i.e. the temperature-dependent resonance integral (3.3). Using the abbreviations

 $N \sigma_{\mathbf{p}}' = \Sigma_{\mathbf{p}}' = \Sigma_{\mathbf{p}} + \mathbf{S}$  $N \sigma_{\mathbf{t}}' = \Sigma_{\mathbf{t}}' = \Sigma_{\mathbf{t}} + \mathbf{S}$ 

ß

 $x = \frac{2}{r} (E - E_p).$ 

 $I = \sum_{\mathbf{g}} \frac{\sigma_{\mathbf{p}}' \Gamma_{\mathbf{\gamma}}}{\Delta E \cos 2\delta_{1}} J(\mathbf{e}^{\mathbf{g}}, \mathbf{B}^{\mathbf{g}})$ (3.6)

with

$$s = \frac{\Delta}{\sigma_{p}}$$

$$s = \frac{\sigma_{p}}{\sigma_{oc}} \cos 2\delta_{1} s$$

**r**s

Because the nuclear resonances are very narrow the energy dependence of  $\theta^s$  and  $\beta^s$  are ignored over the range of integration. Besides this, it has to be summed over the different resonance series (index s).

In the statistical resonance region the resonance integral (3.6) has to be averaged over the Porter-Thomas statistical chi-square  $\Gamma_n$  distribution of one degree of freedom. The radiative capture width  $\Gamma_y$  is taken to be constant. We did this averaging by following a method described in Ref. [7].

Finally, the energy range of integration  $\Delta E$  must be chosen. So far we have assumed only one resonance in  $\Delta E$ . In reality we had n<sup>6</sup> resonances in  $\Delta E$  owing to the limited energy resolution of the neutron spectrometer.

With the aid of

$$\frac{\Delta E}{n} = D^{S}$$
(3.7)

Eq.(3.6) can be rewritten:

I

$$= \int_{\mathbf{s}} \frac{\sigma_{\mathbf{p}} \cdot \Gamma_{\gamma}}{\overline{\mathbf{p}^{\mathbf{s}} \cos 2\delta_{1}}} J(\mathbf{e}^{\mathbf{s}}, \mathbf{\beta}^{\mathbf{s}})$$
(3.8)

The average energy dependent level spacing was calculated from the average level density.

#### 3.3. A code for the calculation of the resonance integral

A code has been written for the IBM 7074 to calculate hot-to-cold foil activation ratios containing as an option hot-to-cold resonance integral ratios. This code computes the hot-to-cold ratios using the formulas and averaging methods mentioned in sections 3.1 and 3.2. No resonance overlap corrections have been applied. The basic nuclear data, which are incorporated into the code for  $^{238}$ U, are taken from Schmidt [8] and given in Table III.

The tabulated J-integral was used except for small  $\theta$  and  $\theta/\beta$  values for which Dresner [4] has given a power series yielding an accurate solution. Computer running time for one ratio at one energy is 8 minutes.

1	2	3
	4.65×10 <sup>6</sup>	
20.8	20.8	11.4
	24.8	
1.87	5.2	2.85
	9.18	
	1 20.8 1.87	1 2 4.65×10 <sup>6</sup> 20.8 20.8 24.8 1.87 5.2 9.18

#### TABLE III. RECOMMENDED NUCLEAR DATA FOR <sup>238</sup>U

#### 4. DISCUSSION AND CONCLUSIONS

The investigations described have demonstrated the possibility of using the slowing-down time spectrometer in the energy range from 0.2 to 30 keV to measure the Doppler effect of heated samples. For these experiments the limited energy resolution of the spectrometer is advantageous because averaging over  $\Delta E$  is necessary to compare experi-

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ment with theory in the statistical energy region. The experiments described have to be taken as a first step in examining the capabilities of the slowing-down time spectrometer for this type of investigations. Although sufficiently small error margins have been obtained, for instance, for tantalum, an improvement is necessary for materials with low signal-to-background ratios, e.g.  $^{238}$ U. This improvement can certainly be achieved by using faster electronics to decrease deadtime effects. From the results for  $^{238}$ U obtained so far, it can be concluded that an accuracy of the measured ratio of about 5 to 10% seems possible.

Further, it can be concluded from the experience gained so far that other interesting aspects of the Doppler effect can be investigated by this technique. These aspects are: overlap and interference effects of different resonance series and different nuclides; the low energy extension to which the statistical resonance theory holds; and the saturation range of the Doppler effect at high temperature.

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#### NOMENCLATURE

- A Atomic mass (mass units)
- $\delta_1$  Phase shift for neutrons with quantum number 1
- $\overline{D^s}$  Mean level spacing of series s (eV)
- $\Delta$  Doppler width (eV)
- E Energy (eV)

g	94		SEUFERT and STEGEMANN
Z	ΔE	-	Energy range (eV)
Ī	Ē	-	Mean energy (eV)
F	E <sub>R</sub>	-	Resonance energy (eV)
I	Г	-	Total width (eV)
I	Γ,	-	$\gamma$ -width (eV)
Ī		-	Neutron width of series s (eV)
I	[	-	Effective resonance integral (barns)
J	J	-	J-integral
j	j.	-	Spin quantum number of compound nucleus
l	Ī	-	Mean chord length (cm)
λ	۱s	-	Scattering mean free path (cm)
1	L	-	Angular momentum quantum number of compound nucleus
n	n <sub>N</sub>	-	Neutron mass (mass units)
ľ	N	-	Density of sample material (cm <sup>-3</sup> )
I	P,	-	Escape probability
Ī	R	-	Hot-to-cold $\gamma$ -ray ratio
S	3	-	Number of resonance series
σ	<sup>7</sup> a	-	Microscopic absorption cross-section (barns)
σ	<sup>7</sup> Р	-	Potential cross-section (barns)
C	, t	-	Total cross-section (barns)
a	7 <sub>00</sub>	-	Compound nucleus cross-section at $E = E_R$ (barns)
c	J <sub>oa</sub>	-	Absorption cross-section at $\mathbf{E} = \mathbf{E}_{\mathbf{R}}$ (barns)
Σ	Σ <sub>p</sub>	-	Macroscopic potential cross-section (cm <sup>-1</sup> )
Σ	Σt	-	Macroscopic total cross-section (cm <sup>-1</sup> )
	S	-	Summation symbol
t	t	-	Slowing-down time (µs)
-	Г	-	Temperature (°K)
7	V	-	Volume (cm <sup>3</sup> )
Ģ	þ	-	Flux in $\Delta E \ (cm^{-2}s^{-1})$
q	p	-	Flux per energy unit $(cm^{-2}s^{-1}eV^{-1})$ .

## PREDICTION OF NEUTRON CROSS-SECTIONS IN THE ENERGY RANGE 1.0 TO 10.0 MeV

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#### Abstract

PREDICTION OF NEUTRON CROSS-SECTIONS IN THE ENERGY RANGE 1.0 TO 10.0 MeV. Neutron cross-sections, total, 'shape elastic' and 'reaction', in the energy range 1.0 to 10.0 MeV have been calculated for niobium, lead, uranium-235 and plutonium-239 – which are extensively used in fast nuclear reactors as structural, shielding and fissile materials – by means of the local spherical optical model. It is concluded that highly deformed nuclides like uranium-235 and plutonium-239 can be represented fairly well with a suitable local spherical optical potential and the cumbersome approach of the non-local deformed optical model can be avoided in the first approximation. The real part (U) of the neutron-nucleus potential is represented by the Woods-Saxon form and the imaginary part (W) by the Gaussian form. The whole range of 1.0 to 10.0 MeV has been split up into slabs over which the average values of U, W, a (surface diffuseness), b (Gaussian width) and  $\gamma_0$  have been determined which give good fits to the measured total cross-sections. The maximum deviation from the measured value is found to be 15% but in general the agreement is reasonably good. For niobium no measurements are available beyond 3.0 MeV and so these calculations serve to fill in the gap. For the doubly magic nuclide lead the imaginary part of the potential is small as expected. The reaction cross-section has been split up into compound elastic and inelastic parts by using the Hauser-Feshbach statistical theory.

#### INTRODUCTION

The phenomenological approach to the optical model [1, 2] is being widely used to understand the nucleon-nucleus interactions for heavy and medium-weight nuclei both in the intermediate and high energy regions. It has met with great success in describing the neutron-nucleus interactions which are important in the design of nuclear power reactors and has provided a mechanism of interpolating or extrapolating the data from the measured quantities like total and elastic scattering cross-sections. The optical model is an intermediate between the continuum model and the shell model of the nucleus. If the imaginary part of the potential becomes zero, the nuclear shell model is obtained. If the ratio between the imaginary and real part of the potential tends to infinity, the continuum model results. It determines the 'shape elastic' and 'reaction' cross-sections. The latter is split up into compound elastic and inelastic components using the Hauser-Feshbach statistical theory [3]. Thus:

$$\sigma_t = \sigma_{se} + \sigma_r$$
 (optical model) (1)

$$\sigma_r = \sigma_{ce} + \sigma_{inel}$$
 (statistical theory) (2)

To calculate inelastic scattering cross-sections precise knowledge of the energy levels of the target nucleus, their spins and parities is desired. Any gap in the knowledge of levels or any uncertainty in their spins and parities would mar the calculations. Angular distributions of

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elastically and inelastically scattered neutrons are also predicted using these two models and then one can obtain  $\overline{\mu}$  – the average cosine of the scattering angle in the laboratory system according to the relation:

$$\overline{\mu} = \frac{\int \sigma_{el}(\theta) \mu d\Omega}{\int \sigma_{el}(\theta) d\Omega}$$
(3)

The transport cross-section which governs the leakage of neutrons from the reactor and thus plays a key role in reactor analysis can be obtained as:

$$\sigma_{\rm tr} = \sigma_{\rm t} - \overline{\mu} \, \sigma_{\rm el} \tag{4}$$

In the optical model analysis the Schrödinger equation is numerically solved to obtain  $\eta_{lj}$  — the relative amplitude of the outgoing wave with the following complex potential:

$$V(r) = -Uf(r) - iWg(r) - V_{eo}h(r)\ell \sigma$$
(5)

where

$$f(\mathbf{r}) = \left[1 + \exp\left(\frac{\mathbf{r} - \mathbf{R}}{a}\right)\right]^{-1} \qquad (Woods-Saxon form)$$
$$g(\mathbf{r}) = \exp\left[-\left(\frac{\mathbf{r} - \mathbf{R}}{b}\right)^{2}\right] \qquad (Gaussian form) \qquad (6)$$

and

$$R = \gamma_0 A^{1/3}$$

 $h(\mathbf{r}) = \left(\frac{\hbar}{\mu_{\pi}c}\right)^2 \frac{1}{\mathbf{r}} \frac{df(\mathbf{r})}{d\mathbf{r}}$ 

The transmission coefficients  $T_{\ell j}$ , 'shape elastic' and 'reaction' cross-sections are then given by:

$$T_{\ell j} = 1 - \left| \eta_{\ell j} \right|^2 \tag{7}$$

$$\sigma_{se}(\theta) = \pi \chi^2 \left[ \sum (2\ell+1)(1-\eta_{\ell j}) \frac{P_{\ell}(\cos \theta)}{\sqrt{4\pi}} \right]^2$$
(8)

$$\sigma_{\mathbf{r}} = \pi \chi^2 \sum \left( 2\ell + 1 \right) T_{\ell j} \tag{9}$$
On the basis of Hauser.-Feshbach theory [4] the inelastic cross-section and its angular distribution are given by:

× W(Jj'Jj', I'L) W(JjJj, IL) P<sub>L</sub> (cos  $\theta$ )

(11)

where

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 $I_0$  is the spin of the target nucleus;  $L \le min.$ (2l, 2l', 2J) Z (abcd, ef) are the Z-coefficients of Blatt and Biedenharn [5] W(abcd, ef) are the Racah coefficients [6] Compound elastic cross-section is obtained by letting E = E',  $I_0 = I_a$ , in Eqs (10) and (11).

# **CALCULATIONS**

In the present work computations have been made of neutron crosssections for niobium, lead, uranium-235 and plutonium-239, which are frequently used in nuclear power reactors as structural, shielding and fissile materials, in the energy range 1.0 to 10.0 MeV with the ABACUS-2 Code [7]. The total cross-sections for these materials have been taken from BNL-325 [8] and KFK-120 [9]. To fit these cross-sections optical parameters have been determined by breaking the complete range of 1.0 to 10.0 MeV into several slabs over which the parameters do not show any appreciable energy dependence. Scanning of optical parameters to give good fit at each energy point is possible but it is more laborious and time-consuming. Moreover, it is not required when the average nuclear behaviour of several nuclides having similar nuclear structure such as the nuclear size and shape, the texture of surface, the effects of closed shells and of nuclear spin, are intercompared.



FIG.1. Total cross-sections for niobium.

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The cross-section predictions contained in this paper have been done with the local spherical optical model with the Woods-Saxon form for the real part of the optical potential. To allow for the surface absorption of neutrons the Gaussian form of the imaginary potential is included. This form of potential could give reasonable estimates for the total crosssections and thus no need was felt to include mixed volume and surface interactions or volume interactions alone. Moreover, the projectile neutron meets a sufficient number of nucleons on the surface of the nuclides included in this work and thus the probability of its interacting with these nucleons is greater than with those contained in the inner core.

The spin-orbit term has been dropped since no attempt was made to fit the differential elastic cross-sections and polarization, the data being very meagre, especially for Nb, <sup>235</sup>U and <sup>239</sup>Pu; although for reasonably good estimates of local parameters the data on angular distributions, polarization, inelastic scattering and total cross-sections should, as far as possible, be fitted simultaneously. Nevertheless the transmission coefficients obtained with the local optical parameters scanned here and used in the predictions of inelastic scattering cross-sections on the basis of the Hauser-Feshbach theory do provide orders of magnitude. These orders of magnitude may be of some use in reactor calculations since data on inelastic cross-section measurements for these materials are not available throughout the whole energy range considered here.

#### NIOBIUM

Here the advantage has been taken of an earlier work [10] in which optical parameters for zirconium were scanned in the energy range 1.0 to 10.0 MeV. Zirconium contains 40 protons and 50 neutrons while niobium consists of 41 protons and 52 neutrons. Their average nuclear behaviour is expected to be similar since they are of the same size and shape. The optical potential parameters which have been used in the predictions are:

(1)  $1.0 \le E < 2.0 \text{ MeV}$ ; U = 40 MeV; W = 10 MeV; a = b = 0.5 fm

 $\gamma_0 = 1.555 \text{ fm}.$ 

(2)  $2.0 \le E \le 10.0$  MeV; U = 40 MeV; W = 10 MeV; a = b = 0.6 fm

 $\gamma_0 = 1.25 \text{ fm}.$ 

Between 1 and 2 MeV the value of the cut-off radius is noted to be more than is required for the rest of the incident neutron energies. This is justified since the increased cut-off radius would include even those compound nuclear levels which have  $\Gamma \sim D$  and thus the average of total cross-section taken over all these levels is improved. If the number of compound nuclear levels is inadequate, the measured total cross-section and the calculated one would show disagreement. A comparison of the measured and the calculated data is shown in Fig.1. The maximum deviation of -16% is noted at a neutron incident energy of 2.0 MeV but beyond 2.0 MeV the agreement is complete. No measured data are

E(MeV)	σ <sub>t</sub> (measured)	σ <sub>t</sub> (calculated)	σ <sub>se</sub>	σ <sub>r</sub>
1.0	7.10	7.25	5.32	1.93
2.0	5.10	4.35	2.67	1.67
2.5	4.60	4.46	2.99	1.47
, 3.0	4.30	4.42	3.19	1.23
3.5	-	4.29	3.27	1.02
4-0	-	4.14	3.29	0.85
4.5	-	3.98	3.25	0.73
5.0	-	3.83	3.18	0.65
5.5	-	3.68	3.09	0.59
6.0	· -	3.53	2.97	0.56
6.5	-	3.40	2.85	0.55
7.0	-	3.29	2.72	0,57
7.5	. <b>-</b>	3.21	2.59	0.62
8.0	-	3.16	2.47	0.69
8.5	-	3.12	2.36	0.76
9.0		3.07	2.26	0.81
9.5	-	2.97	2.15	0.82
10.0	-	2.86	2.06	0.80

TABLE I.	TOTAL, SHAPE	ELASTIC AND	REACTION	CROSS-SECTIONS	(BARNS) OF	NIOBIUM

available beyond 3.0 MeV (BNL-325), so these calculations serve to extend it up to 10.0 MeV. The total, shape elastic and reaction cross-sections are given in Table I.

To break the reaction cross-section into compound elastic and inelastic components the following energy levels with their spins and parities have been used:

Energy level (MeV)	J <sup>#</sup>
0.0	9/2+
0.029	1/2
0.741	$7/2^{+}$
0.809	$5/2^{+}$
0.958	9/2+
1.08	$5/2^{+}$

Compound elastic and inelastic cross-sections have been calculated at 2.0, 4.0, 6.0, 8.0 and 10.0 MeV with the NEARREX Code [11], which accounts for level width fluctuation correction, and are recorded in Table II. Any excitation of levels due to direct interaction is not accounted for. Angular distributions of elastically scattered neutrons at 4.0 and 5.0 MeV were also calculated with the ABACUS-2 code to compare them with those measured by Thomson [12] at 90°. The comparison is as follows:

E(MeV)	Thomson's data	Calculated data
4.0	$83 \pm 16 \text{ mb/sr}$	72  mb/sr
5.0	$72 \pm 14 \text{ mb/sr}$	82  mb/sr

The calculated quantities are within the experimental errors.

#### LEAD-208

Lead-208 contains 82 protons and 126 neutrons. Thus it is a doubly magic nuclide with closed shells. This property should result in low values of the imaginary potential, as has been found.

The total cross-sections are taken from BNL-325 and they are the total cross-sections for natural lead. Since natural lead consists mostly of  $^{208}$ Pb, the total cross-sections calculated for this isotope are compared with those of natural lead in Fig. 2.

The surface diffuseness parameter a and the Gaussian width parameter b are fixed at 0.5 fm and 0.8 fm respectively throughout the whole energy range. The value of  $\gamma_0$  has been taken to be 1.25 fm. The real and imaginary parts of the potential vary as follows:

(1)  $1.0 \le E < 2.0 \text{ MeV}$ ; U = 40 MeV; W = 3 MeV

(2)  $2.0 \le E \le 3.5$  MeV; U = 45 MeV; W = 3 MeV

(3)  $3.5 \le \le 5.5$  MeV; U = 55 MeV; W = 3 MeV

(4)  $5.5 \le 10.0 \text{ MeV}$ ; U = 55 MeV; W = 5 MeV

E(MeV)	Compound	Total	Excited levels (MeV)							
	elastic	melastic	0.029	0.741	0.809	0.958	1.08			
<u></u>										
2.0	0.822	0.819	0.004	0.277	0.141	0.312	0.085			
4.0	0.241	0.595	0.007	1.156	0.086	0.259	0.087			
6.0	0.169	0.377	0.017	0.079	0.055	0.154	0.054			
8.0	0.238	0.440	0.031	0.116	0.072	0.157	0.064			
10.0	0.213	0.562	0.031	0.144	0.097	0.198	0.092			
·····	L			<u> </u>	·					

# TABLE II. CALCULATED CROSS-SECTIONS (BARNS) FOR NEUTRON EXCITATIONS OF NIOBIUM



FIG.2. Total cross-sections for lead.

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E(MeV)	$\sigma_t$ (measured)	$\sigma_t$ (calculated)	σ <sub>se</sub>	σ <sub>r</sub>
1.0	6.20	6.28	6.05	0.23
1.5	5.30	5.74	5.53	0.21
2.0	5.70	6.11	4.99	1.21
2.5	6.80	6.98	5.75	1.23
3.0	7.40	7.02	6.11	0.91
3.5	7.60	6.30	5,61	0.69
4.0	7.70	5.93	5.07	0.86
4,5	7.40	6.42	5.54	0.88
5.0	7.20	6.62	5.83	0.80
5.5	6.80	6.57	5.88	0.68
6.0	6,20	6.14	5.32	0.82
6.5	5.80	5.90	5.18	0.72
7.0	5,60	5.64	4.99	0.65
7.5	5.10	5.38	4.78	0.60
8.0	4.90	5.13	4.55	0.58
8.5	4,65	4.92	4.33	0.59
9.0	4,50	4.77	4.13	0.64
9.5	4.60	4.73	3.98	0.75
10.0	4,50	4.81	3.90	0.91

TABLE III. TOTAL, SHAPE ELASTIC AND REACTION CROSS-SECTIONS (BARNS) OF I
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From Fig. 2 it may be inferred that the measured and total crosssections agree fairly well except for the region 3.5 MeV to 4.5 MeV. The maximum deviation is observed to be -25% at 4.0 MeV but it falls to -16% at 3.5 MeV and -11% at 4.5 MeV. To obtain better agreement U and W were changed but no success was achieved. However, no scan was made on all the optical parameters to explain this local effect although it is felt that an agreement can be struck with suitably chosen parameters. The calculated total, shape elastic and reaction cross-sections are given in Table III.

The energy levels used in the computations of compound elastic and inelastic cross-sections are as follows:

Energy level (MeV)	<u>J"</u>
0.0	0+
2.615	3-
3.198	5 -
3.475	4 -
3.709	5 -
3.750	3 -
3,960	6 -
4.300	4+

Both the Hauser-Feshbach statistical theory and Moldauer's statistical theory of nuclear collision cross-sections [13] have been used in predicting the compound elastic and inelastic cross-sections. It has been observed that with Porter-Thomas distribution assumed for the partial widths the inelastic cross-sections obtained with Moldauer's theory show reductions as much as a factor of two from those calculated with the Hauser-Feshbach theory — a situation which has been explained by Dresner [14] and Moldauer [15]. It also indicates the uncertainties that may be present in the transmission coefficients and consequently in the optical model parameters which ought to be obtained with the in-elastic scattering data taken into account, since the imaginary potential is closely related to neutron absorption.

In obtaining the optical parameters the data on inelastic scattering measured by Towle and Gilboy [16] were not considered but only the total cross-sections were fitted. The inelastic cross-sections calculated by us are, therefore, not registered here.

# URANIUM-235 AND PLUTONIUM-239

The two fissile materials  $^{235}$ U and  $^{239}$ Pu are the backbone of nuclear industry and as such their study needs great care. All the available information on their nuclear structure has to be carefully examined before applying a model to calculate their interactions with neutrons. It is well known that these two nuclides are highly deformed ones and they must be studied with the non-local deformed optical model of Perey and Buck [17]. But to avoid the cumbersome approach of this model the spherical local optical model has been used to calculate their total cross-sections. The measured and calculated total cross-sections represented in Figs 3 and 4



FIG.3. Total cross-sections for uranium-235.

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FIG.4. Total cross-sections for plutonium-239.

 $\sigma_t$  (calculated) E(MeV)  $\sigma_{\rm f}$  (measured)  $\sigma_{se}$  $\sigma_r$ 6.55 1.41 1.0 6.33 5**. 1**2 1.5 6.67 7.78 5.77 2.01 2.0 7.10 8.08 6.24 1.84 7.62 2.5 7.57 6.21 1.41 7.08 6.33 0.75 3.0 7.78 3.5 7.82 7.18 5.77 1.41 4.0 7.72 7.02 5.94 1.08 6.85 0.86 4.5 7.57 5.99 7.39 6.67 5,96 0.71 5.0 5.5 7.17 6.48 5.87 0.61 6.28 0.55 6.0 6,93 5.73 6.5 6.72 6.08 5.55 0.53 6.54 5.93 5.36 0.57 7.0 6.16 0.82 7.5 6.39 5.34 6.28 6.29 5.54 0.75 8.0 8.5 6.17 6.32 5.64 0.68 9.0 6.08 6.28 5.66 0.62 9.5 6.01 6.19 5.62 0.57 0.55 10.0 5,96 6.09 5.54

TABLE IV. TOTAL, SHAPE ELASTIC AND REACTION CROSS-SECTIONS (BARNS) OF URANIUM-235

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reveal that this kind of approach is suitable at least in the first approximation so long as one does not try to explain inelastic scattering data where rotational and vibrational excitations are important; although local potentials sometimes reproduce the inelastic data completely.

The maximum deviation from the measured data for both  $^{235}$ U and  $^{239}$ Pu has been found to be 15%. The contributions of partial neutron waves up to l = 9 have been taken into account. The optical parameters for these materials are as follows.

#### Uranium-235

The values of W, a, b and  $\gamma_0$  have been fixed at 6.0 MeV, 0.5 fm,0.5 fm and 1.25 fm respectively throughout the whole energy range. The values of U are:

(1)  $1.0 \le E \le 2.0 \text{ MeV}$ ; U = 41.2 MeV

(2)  $2.0 < E \le 3.0 \text{ MeV}$ ; U = 42.5 MeV

(3)  $3.0 < E \le 7.5$  MeV; U = 52.5 MeV

(4)  $7.5 \le 10.0 \text{ MeV}$ ; U = 60 MeV

#### Plutonium-239

The values of W, a, b and  $\gamma_0$  are fixed at 6.0 MeV, 0.5 fm, 0.5 fm and 1.25 fm respectively. The values of U are:

(1)  $1.0 \le E < 1.5 \text{ MeV}$ ; U = 41.2 MeV

(2)  $1.5 \le E \le 2.0 \text{ MeV}$ ; U = 43.5 MeV

(3)  $2.0 < E \le 3.0$  MeV; U = 42.5 MeV

(4)  $3.0 \le E \le 6.5$  MeV; U = 51 MeV

(5)  $6.5 < E \le 10.0$  MeV; U = 60 MeV

These parameters are preliminary and are to be modified in the light of inelastic scattering and angular distribution data.

Excitation cross-sections for 22 energy levels in the case of  $^{235}$ U and for 24 energy levels in the case of  $^{239}$ Pu as suggested by Schmidt [9] were calculated. The compound elastic cross-section was found to decrease as the energy of the incident neutron increased.

The calculated cross-sections for  $^{235}$ U and  $^{239}$ Pu are shown in Tables IV and V respectively.

E(MeV)	σ <sub>t</sub> (measured)	$\sigma_t$ (calculated)	ose	σ <sub>r</sub>
1.0	7.11	7.22	5.18	2.04
1.5	6.75	7.06	5.78	1.28
2.0	6.91	7.09	6.25	0.84
2.5	7.63	7.57	6.71	0,86
3.0	8.0	6.98	6.33	0.65
3.5	8.0	7.33	5.82	1.51
4.0	7.96	7.46	6.20	1.26
4.5	7,91	7.32	6.31	1.01
5.0	7.86	7.08	6.25	0.83
5.5	7.76	6.80	6.11	0.69
6.0	7.62	6.51	5.90	0.61
6.5	7.35	6.22	5.66	0.56
7.0	7.10	6.31	5.41	0.90
7.5	6.85	6.43	5.63	0,80
8.0	6.60	6.45	5.74	0.71
8.5	6.44	6.41	5.76	0.65
9.0	6.33	6.33	5.74	0.59
9.5	6.25	6,23	5.67	0.56
10.0	6,15	6.14	5,58	0,56

TABLE V. TOTAL, SHAPE ELASTIC AND REACTION CROSS-SECTIONS (BARNS) OF PLUTONIUM-239

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# DISCUSSION

T.D. BEYNON: I would be interested to hear from Mr.Rastogi whether the oscillations in the calculated  $\sigma_t$  curves in his paper, as exemplified by Fig.2 for lead, are due to the energy limits in the averaging procedure used for the optical model calculations or to the target-level scheme that he applied? Also, could we please have some idea of the errors on the experimental points used for the comparison?

B.P. RASTOGI: We tried to remove the oscillations by changing U and W but were unsuccessful. No scan was made on any other parameters to explain this effect but it is felt that agreement could be achieved with suitably chosen parameters. Furthermore, in our potential there is no spin-orbit term; if this were introduced it might well lead to enhanced agreement with the experimental points.

# SELF-SHIELDING FACTORS FOR URANIUM AND PLUTONIUM

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#### Abstract

SELF-SHIELDING FACTORS FOR URANIUM AND PLUTONIUM. Self-shielding factors for <sup>238</sup>U, <sup>239</sup>Pu, <sup>240</sup>Pu, <sup>241</sup>Pu and <sup>242</sup>Pu are presented for several energies and temperatures as functions of dilution. They were calculated by the 'ladder' method, in a variant which was especially developed to gain high accuracy in short computer time. A brief description of the method is given.

• The temperature-dilution dependence of the cross-sections displayed by these shielding factors is thus free from the assumptions of either small fluctuations or isolated resonances.

#### INTRODUCTION

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In the calculation of the Doppler effect in fast reactors it is useful to introduce the concept of effective cross-sections. An effective crosssection for the energy E is the weighted average of the resonance crosssection in a small energy range  $\Delta E$  about E, the flux being the weight function. A microscopic effective cross-section for an element of the mixture depends on the mixture characteristics, which, in a common approximation [1], are the background cross-section  $\sigma$  and the mixture temperature T. If the range  $\Delta E$  is narrow enough to permit the collision density F(u) to be considered a constant, and if, further,  $\Delta E \ll E$ , so that the factor 1/E can be neglected in the averaging process, the effective cross-section is of the following form:

$$<\sigma_{\mathbf{x}} > (\mathbf{E}, \sigma, \mathbf{T}) = \frac{\int_{\mathbf{A}}^{\mathbf{A}\mathbf{E}} \frac{\sum_{\mathbf{i}} \sigma_{\mathbf{x}, \mathbf{i}}^{\mathbf{o}} \psi\left(\frac{\mathbf{E}-\mathbf{E}_{\mathbf{i}}}{\Gamma_{\mathbf{i}}}, \mathbf{T}\right)}{\int_{\mathbf{A}\mathbf{E}} \frac{\mathbf{A}\mathbf{E}}{\sigma + \sum_{\mathbf{i}} \sigma_{\mathbf{t}, \mathbf{i}}^{\mathbf{o}} \psi\left(\frac{\mathbf{E}-\mathbf{E}_{\mathbf{i}}}{\Gamma_{\mathbf{i}}}, \mathbf{T}\right)}$$
(1)

In Eq.(1), x is the type of reaction, i the resonance index,  $\sigma^{\circ}$  the cross-section at the resonance peak,  $\psi$  the line shape, and  $\Gamma$  the total resonance width.  $\sigma_{x,i}^{\circ}$  depends further on the neutron width  $\Gamma_{n,i}$ , and on the width  $\Gamma_{x,i}$  for the reaction x. The summation over i includes, strictly, every resonance in the range from 0 to some cut-off energy  $E_c$  where the resonance structure disappears. In the range of low energies, however, the average width or Doppler width of the resonance is much smaller than the average resonance spacing, so that each  $\langle \sigma_x \rangle$  (E,  $\sigma$ , T)

Isotope	Last resolved resonance (eV)	Number of resonances	Reference
U-238 Pu-239	4000	239 87	SCHMIDT [10]
Pu-240	680	43	naper SM-101/21
Pu-241 Pu-242	63 390	6 <b>1</b> 20	(these Proceedings)
<u> </u>			

# TABLE I. THE KNOWN PART OF THE RESONANCE LADDER AS OF JANUARY 1967

is determined in practice by one resonance, namely the one closest to E. The number of i terms will increase as the energy increases due to increased overlapping between resonances.

The evaluation of an effective cross-section is straightforward so long as the energy and widths of each of the resonances in the summation in Eq.(1) are known. However, considering that the Doppler range is from 100 eV to 100 keV, Table I shows that only a small fraction of the resonances concerned are actually known.

To calculate the necessary effective cross-sections one has, therefore, to rely heavily on approximate methods. In doing so three assumptions have been made: firstly, that the level spacing and the resonance parameters are statistically distributed about mean values; secondly, that the level spacing has a Wigner distribution and the widths a  $\chi^2$ -type distribution with an a priori unspecified number  $\nu$  of degrees of freedom [2, 3]; and thirdly, that the mean parameters and their energy dependence are known.

Until recently, two analytic methods were known for the evaluation of effective cross-sections in the range of unresolved resonances [2, 3]. In the one, applicable to the low energy range, it is assumed that the cross-section (Eq. (1)) is determined everywhere by one resonance – the nearest. In the other, applicable to the high energy range, it is assumed that resonance overlapping makes the cross-section fluctuations small. There exists, however, an intermediate range, approximately from 500 eV to 10000 eV, where neither method is sufficiently reliable since the resonances are neither spaced enough for the overlapping to be negligible nor close enough for the overlapping to be strong.

In an attempt to avoid assumptions concerning the extent of resonance overlapping, a statistical-numeric method has lately been devised for the evaluation of Eq.(1) [4-6]. The essence of the method is the construction of an artificial distribution for each resonance parameter  $z_i$  in the range  $\Delta E$ . This distribution, called a "ladder", is constructed by repeated sampling of a random number between 0 and 1. Each random number

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determines a value of  $z_i$  from the corresponding statistical distribution  $y_i(z_i)$ . Values of  $z_i$  obtained in consecutive random samplings are assigned to consecutive resonances. The use of the random distribution creates a population of effective cross-sections  $\langle \sigma \rangle_{\ell}$ , each obtained by a different ladder  $\ell$ , and the mean  $\langle \langle \sigma \rangle_{\ell} \rangle$  will be a good estimate of the cross-section built on the real ladder,  $\langle \sigma \rangle_{\ell}^*$ , to the extent that the distribution of the population is narrow.

One question that arises is how many ladders should be sampled out in order to get a good estimate of the cross-section and its standard deviation. As Kelber and Kier found, and as is also our practice, some 10 to 15 ladders suffice. This has a practical importance since machinetime is proportional to the number of ladders used to evaluate a crosssection.

The second question is how many resonances to include in a ladder such that the statistical error be as low as required. The practice of Dyos and Stevens was that large ladders of up to a thousand resonances should be considered in order to get a few percent accuracy in the crosssection and that for problems requiring high accuracy the ladders should be much larger. These findings render the procedure very timeconsuming or even unfit as a standard efficient method for the evaluation of effective cross-sections. A further difficulty in this respect is the fact that the ranges  $\Delta E$  for the calculation of the effective cross-sections should be small enough to ensure a constant collision density and will not, in general, contain enough resonances as to obtain a sufficiently low statistical error.

#### A LADDER METHOD BY DISCRETE SAMPLING

To render the ladder method an efficient computer routine, one has to find a way of achieving high accuracies with ladders of a comparatively small number of resonances. If we look into the reason for the statistical dispersion we find it to have two sources. One source of statistical error is the different order in which the parameters are distributed among the resonances, in different ladders. This sort of randomness cannot be repaired: it is the immediate result of the method — the random sampling. Another source for statistical error is the fact that in sampling parameters for a ladder we pick a finite number of values out of a continuous distribution and therefore one ladder will differ from the other not only in the order of the parameters but also in the values of the parameters. If each ladder contained the same parameter values one source for statistical inaccuracy would be removed.

Let y(z) be the distribution of the width or the level spacing, where the range of z is  $0 \le z \le \infty$ . Divide this range into N equi-probability subranges defined by the sequence of points  $0 = z_0 < z_1 < z_2 < \ldots < z_N = \infty$ , such that the following holds:

$$\int_{0}^{2} y(z) dz$$

$$\frac{z_{n-1}}{\sum_{0}^{\infty}} = \frac{1}{N} \quad (\text{for each } n)$$

$$\int_{0}^{0} y(z) dz$$

(2)

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Define "partial" moments of the distribution as follows:

$$< z^{m} >_{n} \equiv \frac{\int_{z^{m}y(z)dz}^{z_{n}}}{\int_{y(z)dz}^{z_{n-1}}} \quad (\text{for each } n)$$
(3)

According to Eqs (2) and (3)

$$\langle z^{m} \rangle \equiv \frac{\int_{0}^{\infty} z^{m} y(z) dz}{\int_{0}^{\infty} y(z) dz} = \frac{1}{N} \sum_{n=1}^{N} \langle z^{m} \rangle_{n}$$
 (4)

Thus the moments of y(z), and particularly the average, are entirely defined by the partial moments  $\langle z^m\rangle_n$  . Under what conditions can we calculate effective cross-sections if the continuous distributions y(z) are replaced by the discrete distributions  $y(\langle z \rangle_n)$ ? It is obvious that the calculation will become more reliable as N increases. Fig. 1 shows that for values higher than N = 6 all the calculations yield practically the same result. The figure illustrates the influence of N on the calculation of the capture cross-section of <sup>240</sup>Pu at 1 keV. A similar picture was obtained in calculating capture and fission cross-sections at other energies and for other isotopes. As regards accuracy, we get a standard deviation of 2% for a ladder of 300 resonances, compared with 4.4% obtained by Dyos. By using discrete distributions it is possible to obtain high accuracy also in ranges  $\Delta E$  where there are not many resonances (as for example in the higher isotopes of Pu, where resonances have not been resolved up to as high energies as in <sup>238</sup>U); this is because I resonances can be described by a distribution of I values and a ladder of  $I \times J$  stages where J can be taken as large as desired.

Average cross-sections, which are effective cross-sections for infinite dilution, have been measured for many more cases than temperature-dependent finite-dilution effective cross-sections. Therefore an efficient method of calculation is needed particularly for the latter. Separate the effective cross-section into two parts as follows:

$$\langle \sigma_{\mathbf{x}} \rangle (\sigma, \mathbf{T}) = \mathbf{f}_{\mathbf{x}}(\sigma, \mathbf{T}) \langle \sigma_{\mathbf{x}} \rangle$$
 (5)



FIG.1. Calculation of a capture cross-section by a ladder of 120 resonances.

where  $\langle \sigma_x \rangle$  is the average cross-section. Eq.(5) is the definition of the self-shielding factor  $f_x(\sigma, T)$ . To calculate the temperature effect in the reactor one needs the differences  $\langle \sigma_x \rangle (\sigma, T_2) - \langle \sigma_x \rangle (\sigma, T_1)$ . According to Eq.(5)

$$\Delta < \sigma_{\downarrow} > (\sigma_{\downarrow}\Delta T) = \Delta f_{\downarrow}(\sigma_{\downarrow}\Delta T) < \sigma_{\downarrow} >$$
(6)

If  $\langle \sigma_x \rangle$  is known, as for example by measurement, one has to evaluate  $f_x(\sigma, T)$  in order to determine the effective cross-section. Since the average cross-section  $\langle \sigma_x \rangle$  is the effective cross-section in infinite dilution at any temperature, we get, according to (5), for each ladder  $\ell$ ,

$$\mathbf{f}_{\mathbf{x}}^{\prime}(\sigma,\mathbf{T})_{\boldsymbol{\ell}} = \frac{\langle \sigma_{\mathbf{x}} \rangle (\sigma,\mathbf{T})_{\boldsymbol{\ell}}}{\langle \sigma_{\mathbf{x}} \rangle (\infty,\mathbf{T})_{\boldsymbol{\ell}}}$$
(7)

The self-shielding factor  $(f_x)_{\ell}$  is the ratio of two effective cross-sections calculated by the same ladder. Both these effective cross-sections (obtained from Eq.(1)) deviate from the true cross-sections in the same algebraic direction, and we may thus expect the dependence of  $(f_x)_{\ell}$  on  $\ell$  to be less than that of the effective cross-sections themselves. Fig.2 shows that the statistical reliability of  $f_{\gamma}(100b, 300^{\circ}K)$  and  $f_{\gamma}(100b, 900^{\circ}K)$ , calculated for <sup>240</sup>Pu at 1 keV with a ladder of 120 resonances, is better



FIG.2. Calculation of a self-shielding factor by a ladder of 120 resonances.

than that of  $\langle \sigma_{\gamma} \rangle$  calculated for the same conditions, as shown in Fig.1. Furthermore the statistical reliability of  $\Delta f_{\gamma}$  between the two temperatures is even higher.

Figs 1 and 2 show also that the evaluations of  $\langle \langle \sigma_x \rangle_{\ell} \rangle$ ,  $\langle f_{x,\ell} \rangle$  and the standard deviations require a rather small number of ladders.

#### THE COMPUTER CODE SELF

The computer code SELF computes self-shielding factors as functions of energy, temperature and background cross-section. The input data are average resonance parameters as functions of energy and the number  $\nu$  of degrees of freedom in the distributions of the partial widths and level spacings. The self-shielding factors are calculated from Eq.(1) (including the antisymmetric line shape for elastic scattering) according to the ladder method as described in the previous section. In each region  $\Delta E$  a decision is made at the start as to which resonances to include in the sum  $\Sigma$  in

Eq.(1). The criterion for deciding to include a particular resonance is that its contribution relative to that of the nearest resonance is greater than a predetermined  $\epsilon$  (usually  $\epsilon = 0.001$ ). The relative contribution is evaluated from average  $\Gamma$  and D values for the line shape at zero and at infinite temperature, taking the greater of the two. The line shape is calculated by the WANL routine, developed at Argonne [8]. It is a rapid and exact routine. Its accuracy was checked by comparison with other exact routines which are more time-consuming [9]. In the present version of the ladder method the value of  $\Delta E$  is unimportant and is determined by the number of resonances in the ladder and their average spacing. The number of resonances in the ladder is determined by the requirements for statistical accuracy. The integration over E is carried out by dividing  $\Delta E$  into subregions and performing a 48-point Gaussian integration within each. The number of sub-regions is determined by the requirement that its aug-

# TABLE II.SELF-SHIELDING FACTORS FOR URANIUM-238 AND PLUTONIUM-239

E	т	c	U-2 <b>3</b> 8 apture		U-238 elastic			Pu-239 capture			Pu fi	<b>-</b> 239 ssion		Pu-239 elastic		
(keV)	( <b>°</b> K)	1000	σ 100	10	1000	σ 100	10	1000	σ 100	10	1000	σ 100	10	1000	σ 100	10
0.1	300	.462	.157	.066	•574	.402	•347	.801	.425	.213	.884	.618	.403	.940	.841	.792
	900	.546	.194	.077	•624	.416	•351	.862	.513	.269	.915	.667	.437	.957	.862	.806
	1500	.590	.218	.085	•653	.427	•355	.887	.564	.308	.928	.696	.462	.965	.875	.815
	2100	.620	.237	.091	•674	.435	•358	.902	.599	.338	.937	.718	.482	.970	.885	.823
0.3	300	.663	•282	.124	.727	.499	.414	.906	•598	.322	•940	.721	.479	•972	.889	.824
	900	.745	•352	.153	.783	.530	.426	.942	•700	.419	•960	.782	.541	•982	.915	.848
	1500	.782	•392	.173	.810	.550	.436	.955	•747	.473	•968	.811	.576	•986	.928	.862
	2100	.805	•422	.188	.828	.565	.443	.962	•776	.510	•972	.829	.602	•989	.937	.872
1.0	300	•902	.616	.361	.920	.729	•599	.981	.871	.653	•986	.904	.728	•995	.965	.913
	900	•937	.702	.438	.945	.775	•632	.990	.924	.761	•992	.938	.799	•997	.979	.937
	1500	•950	.742	.481	.956	.799	•651	.992	.941	.803	•994	.949	.828	•998	.983	.947
	2100	•957	.768	.511	.962	.815	•665	.994	.950	.827	•994	.956	.845	•998	.986	.954
3.0	300	•973	.842	.629	•974	.867	•739	•993	•945	.808	•994	•953	.834	•998	.985	•948
	900	•984	.894	.716	•984	.903	•781	•996	•967	.874	•996	•970	.885	•999	.991	•966
	1500	•988	.915	.757	•987	.919	•803	•997	•974	.898	•997	•976	.906	•999	.993	•972
	2100	•990	.928	.7 <sup>84</sup>	•989	.929	•819	•997	•978	.911	•998	•980	.918	•999	.994	•976
10	300	•993	•950	.827	•993	•951	.865	•998	•986	•939	•998	•987	•944	.999	•996	.982
	900	•997	•972	.895	•996	•970	.903	•999	•992	•963	•999	•992	•966	1.000	•99 <b>7</b>	.988
	1500	•998	•980	.921	•997	•977	.921	•999	•993	•971	•999	•994	•974	1.000	•998	.991
	2100	•998	•984	.935	•998	•980	.932	•999	•995	•976	•999	•995	•978	1.000	•998	.992
40	300 900 1500 2100	.999 .999 1.000 1.000	.991 .996 .997 .998	•957 •980 •986 •989	.999 1.000 1.000 1.000	•992 •996 •997 •997	.966 .981 .986 .988									

E	T	Pu-240 capture		Pu fi	-240 ssion	Pu- ela	240 stic	Pu- ca	241 pture	Pu- fis	241 sion	Pu- ela	241 stic	Pu-2 capt	42 ure	Pu-24 elast	2 ic
(keV)	(°K)	3000	σ 300	3000	σ 300	3000	σ <b>30</b> 0	10000	σ 1000	10000	σ 10 <b>00</b>	10000	σ 1000	م 10000	1000	σ 10000	1000
0.1	300 900 1500 2100	.665 .743 .780 .803	•266 •329 •367 •395	.663 .741 .777 .800	•268 •331 •369 •397	.684 .748 .780 .800	.396 .430 .452 .470	.987 .992 .993 .994	.892 .927 .941 .949	•992 •994 •995 •996	•934 •951 •958 •963	•997 •998 •998 •998 •999	.976 .984 .987 .989	.856 .901 .918 .928	.476 .563 .606 .635	.872 .909 .924 .933	.601 .651 .678 .697
0.3	300 900 1500 2100	.837 .889 .909 .921	•453 •544 •592 •625	.844 .894 .914 .925	.467 .560 .607 .639	.846 .890 .908 .920	.554 .609 .641 .663	• 995 • 997 • 998 • 998	•953 •972 •978 •982	.996 .997 .998 .998	.965 .975 .979 .982	•999 •999 •999 •999 1.000	.989 .993 .995 .996	•937 •959 •967 •972	.665 .745 .782 .805	.944 .963 .970 .974	•731 •785 •812 •829
1.0	300 900 1500 2100	.961 .976 .982 .985	.765 .836 .866 .885	•959 •975 •981 •984	•759 •830 •860 •879	•963 •977 •982 •984	. 798 . 850 . 874 . 889	.999 .999 .999 1.000	.988 .993 .995 .996	•999 •999 1.000 1.000	.992 .995 .996 .996	1.000 1.000 1.000 1.000	•997 •998 •999 •999	.978 .987 .989 .991	.839 .891 .912 .924	.982 .988 .991 .992	.872 .909 .925 .934
3.0	300 900 1500 2100	•99 <b>0</b> •994 •996 •997	.920 .953 .964 .971	.991 .995 .997 .997	.928 .959 .970 .975	•990 •994 •995 •996	.924 .951 .961 .967										
10	300 900 1500 2100	•998 •999 •999 •999	•980 •990 •993 •995	•998 •999 •999 •999	.978 .989 .992 .994	•997 •998 •999 •999	•975 •986 •989 •991										

TABLE III.SELF-SHIELDING FACTORS FOR PLUTONIUM-240, -241 AND -242

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mentation should not result in a relative change larger than 0.001 in the computed quantity. The number of sub-regions needed at low energies (100-1000 eV) is equal to or somewhat greater than the number of resonances in the ladder. For higher energies the number of sub-regions decreases, owing to the increase in resonance overlapping. The code is written for the Philco-2000.

#### DISCUSSION AND RESULTS

Since average cross-sections are usually measured, or at least are much more measurable than effective cross-sections, and since selfshielding factors or differences of self-shielding factors can be calculated to satisfactory accuracy in reasonable computer-time, we find the notion of the self-shielding factor more effective and useful to Doppler calculations than effective cross-sections. As an example we present in the paper self-shielding factors for  $^{238}$ U,  $^{239}$ Pu,  $^{240}$ Pu,  $^{241}$ Pu and  $^{242}$ Pu in several energies, dilutions and temperatures. The values in Tables II and III are with three figures, meaning that differences constructed out of them are significant to the last figure. These tables are part of much more detailed ones calculated as preliminary to the evaluation of group self-shielding factors, for the 30-group set we have constructed. The code written for the Philco-2000 has computed some 6000 self-shielding factors, for energies in the range 100 eV - 50 keV, in 10 hours.

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## DISCUSSION

H.W. KUSTERS: Was a comparison made of the self-shielding of <sup>238</sup>U, as calculated by the ladder method, and the values given by Bondarenko et al., and if so, how large were the deviations in the keV range?

M. SEGEV: Yes, we did do this comparison and found only small deviations, probably due to discrepancies in the resonance parameter data and not to the method of calculation.

H.W. KÜSTERS: Certainly, the Russian set may include other microscopic data, but it is nevertheless of interest to know the deviations.

J.L. ROWLANDS: Could Mr.Segev explain whether he thinks it worthwhile averaging over a number of ladders, when in fact there is only one real ladder? Should we not rather aim at obtaining improved measured data?

M. SEGEV: Naturally, a direct experimental determination of the resonance parameters is preferable, but, failing this, there is no alternative to averaging over a number of ladders. The population of all possible ladders has some standard deviation which indicates the degree of confidence with which we can say that the cross-section value evaluated in this way, as an average, approximates to the actual value.

# BASIC NUCLEAR DATA FOR THE HIGHER PLUTONIUM ISOTOPES

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#### Abstract

BASIC NUCLEAR DATA FOR THE HIGHER PLUTONIUM ISOTOPES. Fast reactor fuel may have an appreciable content of high Pu isotopes, the amount varying according to the source. Therefore, in calculations of static and dynamic reactor characteristics, reliable basic nuclear data are needed not only for <sup>239</sup>Pu but for the higher isotopes as well. Reactor computations depend on cross-sections as functions of energy, dilution and temperature. Basic nuclear data for a fuel isotope consists therefore of average crosssections and resonance parameters. Experiment alone does not furnish such data in a final complete form. In fact, the experimental information needs interpretation, weighting, evaluation and often also interpolation on theoretical grounds. Within the framework of contracted research with the Association EURATOM-Karlsruhe on fast reactors, an evaluation was made of basic nuclear data for the high Pu isotopes. This paper summarizes some pertinent parts and aspects of the evaluation. Resonance parameters and crosssections are presented for <sup>240</sup>Pu, <sup>241</sup>Pu and <sup>242</sup>Pu in the form of experimental and recommended data. Complete sets of parameters include the first 43, 61 and 20 resonances of these isotopes respectively. Average parameters are derived from these sets, to be used at higher energies where either the parametrization is incomplete or the resonances unresolved. Although the samples are, as a rule, too poor for a direct derivation of statistical distributions, there is enough general knowledge on the subject today to fix these distributions within narrow limits.

## 1. INTRODUCTION

Since the plutonium produced as a by-product in thermal reactors is to serve as fuel for fast reactors, it is necessary to have accurate nuclear data for all its isotopes present. The composition of Pu fuel from thermal reactors may be in the following range [1]:  $^{239}$ Pu, 55-60%;  $^{240}$ Pu, 20-25%;  $^{241}$ Pu, 10-15%;  $^{242}$ Pu, 5-10%. That means that if a typical 1000-MW(e) reactor is fuelled with, say, 3 tonnes of plutonium, it will have about 1700 kg  $^{239}$ Pu, 600 kg  $^{240}$ Pu, 400 kg  $^{241}$ Pu and 300 kg  $^{242}$ Pu. In other words, about half or  $1\frac{1}{2}$  tonnes of the plutonium will be of the higher isotopes variety. It is thus advisable to know as much as possible about the nuclear and other properties of half the fuel loading.

Nuclear data for  $^{239}$ Pu have been evaluated recently [2,3]. The purpose of this paper is to give a recommended complete set of nuclear data for the higher isotopes  $^{240}$ Pu,  $^{241}$ Pu and  $^{242}$ Pu.

Only limited experimental data are available, and it was thus necessary to rely heavily on nuclear systematics and theory to cover the whole range of interest (0.01 eV to 15 MeV). The systematic literature search covers the period up to the Paris Conference on Nuclear Data (October 1966), but later available data are also included. The results are presented below according to three energy regions: the thermal region (0.01 to 1 eV approx.), the resonance region, and the fast neutron region (above 1 keV).

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The data evaluated has been punched on cards in the KEDAK (Karlsruhe) format and, like the rest of the Karlsruhe nuclear data file, will be transferred to the neutron data compilation centre at Saclay where it will then become available upon request.

#### 2. THERMAL ENERGY RANGE

In this section we give recommended values for the following crosssections at thermal energy (0.0253 eV): total  $(\sigma_T)$ , scattering  $(\sigma_n)$ , fission  $(\sigma_f)$  and capture  $(\sigma_\gamma)$ . A discussion and graphs depicting the cross-sections in the range from 0.01 eV to the vicinity of the first resonance will be given elsewhere [4].

#### 2.1. Plutonium-240

 $\sigma_{\rm T}$ . We recommend the value  $\sigma_{\rm T} = \sigma_{\rm n} + \sigma_{\rm f} + \sigma_{\gamma} = 281$  b. A measured value of 273 ± 8 b has been reported [5].

 $\sigma_n$ ,  $\sigma_f$ . From a Breit-Wigner calculation using the parameters of the first resonance we obtained the values  $\sigma_n$  = 2.26 b,  $\sigma_f$  = 0.052 b.

 $\sigma_{\gamma}$ . Ten sources mentioned by Drake and Dyos [6] were used to obtain the weighted average  $\sigma_{\gamma} = 279$  b. To obtain agreement with this value a small change was made in  $\Gamma_{\gamma}$  of the first resonance (from  $31 \pm 3$  to  $32.3 \pm 3$  meV).

#### 2.2. Plutonium-241

 $\sigma_{\rm T}$ . We recommend  $\sigma_{\rm T}$  = 1385 ± 20 b, as in BNL-325 (1965) [7]; this value is based on the measurements of Craig and Westcott [8,9] and Simpson and Schuman [10].

 $\sigma_n$ . This was taken from the multilevel calculation of Drake and Dyos [6]:  $\sigma_n = 12.6$  b.

 $\sigma_{\rm f}$ . A recent evaluation by Westcott [11] and measurements by James [12] and White et al. [13] point to values in the range 1012-1022 b; BNL 325 (1965) gives a best value of 950 ± 30 barns, based on older data. We recommend the value of White et al.:  $\sigma_{\rm f} = 1022 \pm 29$  b.

 $\sigma_{\gamma}$ . This value was calculated as  $\sigma_{\gamma} = \sigma_{\rm T} - (\sigma_n + \sigma_f) = 350 \pm 38$  b. The only direct measurement available is a reactor spectrum average by Fields [14]: 390 ± 80 b.

#### 2.3. Plutonium-242

Table I presents the 0.0253 eV data. For  $\sigma_T$  we adopt the weighted average of the measurements of Auchampaugh et al. [18] and Young et al. [19]. It can be seen that the first positive resonance can account for only one third of  $\sigma_T$ . In fact, Young states [20] that he found it necessary to postulate two negative resonances: one at -0.02 eV and another much further away. In the present work we made an approximate fit to  $\sigma_T$  with one negative resonance. The parameters we arrived at are:  $\Gamma_{\gamma}^{-} = 30 \text{ meV}$ ;  $\Gamma_n^{0-} = 0.13 \text{ meV}$ ;  $E_r^{-} = -1.1 \text{ eV}$ . From these parameters we calculated  $\sigma_n$ , and we took  $\sigma_{\gamma} = \sigma_T - \sigma_n$ , obtaining finally:  $\sigma_T = 39.5 \pm 0.8 \text{ b}$ ;  $\sigma_n = 9.0 \text{ b}$ ;  $\sigma_{\gamma} = 30.5 \text{ b}$ .

#### 3. RESONANCE ENERGY RANGE

There has been a great increase in the volume of experimental data in this region during the last two years. If we take as a starting point for the present evaluation Supplement No.2 to BNL-325 dated February 1965 [7], we see that the number of resolved recommended resonance parameters was 9 for  $^{240}$ Pu, 23 for  $^{241}$ Pu and one for  $^{242}$ Pu. In the present report recommended resonance parameters are given for <u>43</u>  $^{240}$ Pu resonances, 61  $^{241}$ Pu resonances and 20  $^{242}$ Pu resonances.

For comparison the following table summarizes the knowledge of resolved resonance parameters for these as well as other nuclides, as of January 1967:

Isotope	Last resolved resonance (eV)	Number of resonances	Reference
<sup>235</sup> U <sup>238</sup> U <sup>239</sup> Pu	150 4000 300	$ \begin{array}{c} 197\\239\\87 \end{array} $	KFK 120 I (Feb. 1966)
<sup>240</sup> Pu <sup>241</sup> Pu <sup>242</sup> Pu	680 63 390	$ \left.\begin{array}{c} 43\\ 61\\ 20 \end{array}\right\} $	Present report

Most of the recent data in this region have been obtained from two sources:

(1) Time-of-flight techniques using the linear electron accelerators of Euratom (Geel, Belgium), Harwell, the Rensselaer Polytechnic Institute and Livermore.

(2) A very important source of new data has become available from time-of-flight measurements preformed using the 1.2 kt nuclear explosion 'Petrel' in Nevada in June 1965 as a neutron source with a neutron yield of  $1.8 \times 10^{23}$  neutrons with a duration of  $0.1 \,\mu$ sec. The nuclear device was located at a depth of 185 m from which a 35-cm diameter pipe provided a flight path for neutrons with samples, solid-state detectors, fission foils and other necessary instrumentation arranged above the surface. In this experiment fission cross-sections of  $^{240}$ Pu,  $^{241}$ Pu,  $^{241}$ Am,  $^{242}$ Am as well as  $^{233}$ U,  $^{235}$ U and  $^{239}$ Pu were measured and analysed.

In the analysis of both recent and old data three kinds of tables were constructed: (a) tables of experimental and recommended resonance parameters (giving all the available and assumed data; (b) tables of resonance parameters for neutronics calculations (giving only those resonances for which all the significant partial widths can be presented);

Reference		Barns	Remarks		
	σγ	σ <sub>n</sub>	σ <sub>T</sub>	1	
Studier et al. [15]	30 <u>+</u> 10			pile neutrons	
Butler et al. [16]	18.6 <u>+</u> 0.8			measured	
BNL 325 (1958) {17]	30 <u>+</u> 2			(?) wrongly assigned to Butler	
Auchampaugh et al. [18]	30 <u>+</u> 2	8.9 <u>+</u> 2.6	38.9 <u>+</u> 1.6	σ <sub>T</sub> measured; σ <sub>Y</sub> from BNL 325 (1958) σ <sub>n</sub> <sup>m</sup> σ <sub>T</sub> - σ <sub>Y</sub>	
Young et al. [19]			39.8 + 1.0	measured	
This report	30.5	9.0	39.5 <u>+</u> 0.8	recommended	

TABLE I. Pu-242 THERMAL DATA

and (c) tables of average resonance parameters. The tables of type (a) (experimental data) have to be presented elsewhere [4] due to their excessive length.

To compute the recommended resonance parameters from the experimental values, we calculated weighted averages and errors. If the error bars did not overlap, a root mean square error was calculated instead. In cases of large discrepancies, some of the measurements had to be rejected.

# 3.1. Plutonium-240

3.1.1. Resonance parameters: values for neutronics calculations (Table II)

Some comments about the source data follow.

The best data are those from the transmission experiment of Böckhoff et al. [21]. The experiments, which used time-of-flight techniques, were performed with the 60-MeV electron linear accelerator of Euratom, which is operated by the Central Bureau for Nuclear Measurements (CNBM), Geel, Belgium. The <sup>240</sup> Pu samples used in the experiment were supplied by the USAEC. Three samples with a total thickness of 0.0675 g/cm<sup>2</sup> were used. The resolution was 6 to 1 nsec/m. In this experiment 158 resonances were found between 20 eV and 5 keV. Between 20 eV and 800 eV  $\Gamma_n^0$  for 36 out of 42 resonances have been determined by shape analysis. The data of Byers et al. [22] were obtained from a nuclear detonation as neutron source. The resolution was 17 to 6 nsec/m. The shape analysis of the data has not yet been published.

Brooks and Jolly [23] investigated the fission cross-section in the 20 to 120 eV range. Their measurements were relative to  $^{235}$ U; fissions were detected by liquid scintillators. Their  $\Gamma_{\rm f}$  are recommended although they are for the most part upper limits. Short descriptions of the measurements from 1963 back to 1955 are included in BNL-325 (1965) [7].

To complete the table of values for neutronics calculations, when  $\Gamma_{\rm f}$  values were missing, it was assumed that  $\Gamma_{\rm f} = \langle \Gamma_{\rm f} \rangle = 0.23 \ {\rm meV}$  (see section 4.1.1).

TABLE II. Pu-240 RESONANCE PARAMETERS: VALUES FOR NEUTRONICS CALCULATIONS

l	Ξ	0,	J	=	$\frac{1}{2}$	for	all	resonances	
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r	E <sub>r</sub> (eV) r	r <sub>n</sub> (meV)	r (meV) Y	<sup>r</sup> f (meV)	Γ <sup>O</sup> (meV)
1	1.056	2,30	32.3	0.006	2.24
2	20.45	2.3	21	0.55	0.51
3	38.33	18	26	0.14	2.91
4	41.64	16.1	33	0.13	2.50
5	66.63	48.0	.47	0.38	5.88
6	72.82	21.8	22	0.49	2.56
7	90.77	13.3	29	0.23	1.39
8	92.50	2.9	32	1.07	0.30
9	105.03	44.2	32	0.75	4.31
10	121.65	13.9	35	1.6	1.26
11	135.22	17.8	38	0.23	1.53
12	151.72	13.6	32	0.23	1.10
13	162.9	8.6	43	0.23	0.67
14	170.3	13.4	37	0.23	1.03
15	186.1	16.0	41	0.23	1.17
16	199.6	0	32	0.23	0
17	239.3	11.3	32	0.23	0.73
18	260.7	25	32	0.23	1.55
19	287.3	125.1	65	0.23	7.38
20	305.1	7.0	32	0.23	0.40
21	318.5	6.0	32	0.23	0.336
22	320.9	18.6	77	0.23	1.04
23	338.7	5.7	32	0.23	0.31
24	346.2	16.2	51	0.23	0.87
25	364.0	30.9	43	0.23	1.62
26	372.3	13.3	32	0.23	0.69
27	405.0	102.5	71	0.23	5.09
28	419.0	8.1	32	0.23	0.396
29	450.2	16.8	102	0.23	0.79
30	466.4	0	32	0.23	0
31	4/3.2	0	32	0.23	0 22
32	494.2	<b>3.1</b>	32	0.23	0.23
27	499.0	20.6	32	0.23	
24	546 8	20.4	32	0.23	1 28
35	553 5	16 7	32	0.23	0.71
37	566 6	30.0	32	0.23	1 26
38	597.2	53.0	32	0.23	2.17
30	608.4	20.5	32	0.23	0.83
40	632.6	12.3	32	0.23	0.49
40	637.8	0	32	0.23	
42	665.5	183	96	0.23	7.11
43	678.9	24.0	32	0.23	0.92

3.1.2. Average resonance parameters below 1 keV (Table III)

The nuclear momentum and parity of <sup>240</sup> Pu is  $I\pi = 0 +$ ; s-wave neutrons lead to states with total angular momentum  $J = \frac{1}{2}$ , while p-wave neutrons lead to  $J = \frac{1}{2}, \frac{3}{2}$ .

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$\langle D \rangle_{J=\frac{1}{2}}^{\ell=0,1} = 13.2 \pm 1.7 \text{ eV}$
$\langle D \rangle_{J=\frac{3}{2}}^{l=1} = 7.26 \text{ eV}$
$\langle \Gamma_n^0 \rangle_{J=\frac{1}{2}}^{\ell=0} = 1.81 \pm 0.24 \text{ meV}$
$\langle \Gamma_n^0 \rangle_{J=\frac{1}{2}}^{\ell=1} = 3.30 \text{ meV}$
$\langle \Gamma_n^0 \rangle_{J=\frac{3}{2}}^{\ell=1} = 1.82 \text{ meV}$
$\xi_{\ell=0}^{J=\frac{1}{2}} = \xi_{0} = (1.37 \pm 0.51) \times 10^{-4}$
$\xi_{\ell=1}^{J=\frac{1}{2},\frac{3}{2}} = \xi_{1} = 2.5 \times 10^{-4}$
$\langle \Gamma_{\rm f} \rangle_{\rm J=\frac{1}{2}}^{\ell=0} = 0.23  {\rm meV}$
$\left\langle \Gamma_{\gamma} \right\rangle_{J=\frac{1}{2}}^{\ell=0} = 32 \pm 1 \text{ meV}$
$\langle \Gamma_{\gamma} \rangle_{J=\frac{1}{2},\frac{3}{2}}^{\ell=1} = 32 \text{ meV}$
$\nu_n = 1;$ $\nu_f = 1;$ $\nu_\gamma = \infty$
σ <sub>pot</sub> = 10.6 barns

TABLE III. Pu-240 AVERAGE RESONANCE PARAMETERS BELOW 1 keV

Average parameters for s-wave neutrons. The integral plot of the resonance levels shows that they begin to be missed at an increasing rate beyond 200 eV. Consequently, the calculation of the average level spacing  $\langle D \rangle$  was based on 15 resonances. We obtained  $\langle D \rangle = (E_{15} - E_1)/14$ = 13.2 ± 1.7 eV. The average reduced neutron width was calculated to 36

be  $\langle \Gamma_n^0 \rangle = (1/36) \sum_{r=1}^{r=1} \Gamma_{nr}^0 = 1.81 \pm 0.24$  meV. The capture width  $\Gamma_{\gamma}$  was

assumed to have a delta function distribution. A weighted average was calculated from the measured  $\Gamma_{\gamma r}(r=1,\ldots,21)$ , namely  $\langle \Gamma_{\gamma} \rangle = 32 \pm 1 \text{ meV}$ . The strength function is  $\xi_0 = \langle \Gamma_n^0 \rangle / \langle D \rangle = (1.37 \pm 0.51) \times 10^{-4}$ . This value may be compared with  $\xi_0 = (2.3 \pm 0.7) \times 10^{-4}$  reported by Simpson and Fluharty [24].  $\langle \Gamma_f \rangle$  was calculated by fitting  $\langle \sigma_f \rangle = 0.16$  b (using the code SELF[25]) with  $\nu_f = 1$  and the present average parameters. The result (which we recommend for the whole 1-1000 eV range) is  $\langle \Gamma_f \rangle = 0.23 \text{ meV}$  at E = 200 eV. The  $\langle \sigma_f \rangle$  was calculated from the resonance curve of Byers et al. [22] in the range 0-1000 eV.

<u>Average parameters for p-wave neutrons</u>. We follow the recommendations of Schmidt for  $^{238}U[2]$ . (D) is assumed to depend on J and not on l, according to the Fermi gas model:

$$\langle D \rangle$$
(J) =  $\frac{\text{const.}}{2J+1} \exp \frac{J(J+1)}{2\sigma^2}$ 

with the spin cut off  $\sigma = 4$ . It follows that:

$$\langle D \rangle_{J=1/2}^{\ell=0, 1} = 13.2 \pm 1.7 \text{ eV};$$
  $\langle D \rangle_{J=3/2}^{\ell=1} = 7.26 \pm 0.94 \text{ eV}$ 

 $\xi_1$  is assumed to be independent of J and equal to 2.5  $\times$  10<sup>-4</sup> (as in  $^{238}$  U). It follows that  $\left<\Gamma_n^0\right>_{J=1/2}^{\ell=1}$  = 2.24 meV, and  $\left<\Gamma_n^0\right>_{J=3/2}^{\ell=1}$  = 1.23 meV.  $\left<\Gamma_\gamma\right>$  is assumed to be independent of J and  $\ell$ . The potential scattering cross-section is taken as that for  $^{238}$ U[2], namely  $\sigma_{pot}$  = 10.6 b.

Errors. The calculated errors are statistical. For a sample  $(x_1, \ldots, x_n)$  from a  $\chi^2$ -distribution with  $\nu$  degrees of freedom, the standard deviation is

$$\sigma = (2/n\nu)^{1/2} \langle x \rangle$$

Statistical distributions. It is accepted at present that D follows a Wigner distribution, while  $\Gamma_n^0$  follows a X<sup>2</sup>-distribution with  $\nu_n = 1$ . The distribution of  $\Gamma_\gamma$  is commonly assumed to be a delta function. The distribution of  $\Gamma_f$  is open to question and, moreover, may depend on the isotope considered. We adopted a X<sup>2</sup>-distribution with  $\nu_f = 1$ .

#### 3.2. Plutonium-241

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3.2.1. Resonance parameters for neutronics calculations (Table IV)

Plutonium-241 presents the problem of level interference. To reproduce the fission cross-section between resonances satisfactorily, it is necessary to use a multilevel multichannel formalism, such as that of Reich and Moore [26]. This has been done by most experimenters. But at present, nuclear codes for Doppler calculations are based on the use of the Breit-Wigner line shapes and therefore the data for neutronics calculations must be presented as B-W parameters. In computing recommended values we did not differentiate between single and multilevel resonance parameters.

The best data for the range 4 to 20 eV come from the fission measurements of Moore et al. [27]. The resolution was 75-100 nsec/m. The best data in the range 20 to 60 eV come from the fission measurements of Simpson et al. [28] who made use of a nuclear detonation as a neutron source; the resolution was 20 nsec/m, and there was also an improvement in the statistical accuracy. Short descriptions of the measurements from 1964 back to 1956 are included in BNL-325 (1965)[7].

TABLE IV.	Pu-241 RESONANCE PARAMETERS:	VALUES
FOR NEUTR	ONICS CALCULATIONS	
$\theta = 0$ for all	*****	

l = 0 for all resonances

r	E <sub>r</sub> (eV)	J	Γ <sub>n</sub> (meV)	Γ <sub>γ</sub> (meV)	Γ <sub>f</sub> (meV)	Γ <sup>ο</sup> (meV)
0	- 0.160	3		40	60	0.0621
1	0.260	3	0.044	35	75	0.0857
2	4.30	3	0.56	40	32	0.27
3	4.58	2	0.52	40	160	0.24
4	5.92	2	2.98	40	1350	1.22
5	6.93	3	0.61	40	94	0.23
6	8.60	3	0.86	40	70	0.28
7	9.5	3	0.15	40	120	0.051
8	10.1	2	1.8	40	900	0.56
9	12.78	2	0.95	40	235	0.26
10	13.40	3	0.19	30	39	0.51
11	14.03	2	0.216	40	214	0.058
12	14.75	2	7.4	31	120	1.92
13	15.98	2	1.82	40	465	0.46
14	16.69	3	1.05	40	180	0.26
15	17.83	3	2.7	33	24	0.65
16	18.2	2.5	0.16	40	27	0.037
17	20.70	3	0.30	40	35	0.066
18	21.05	3	0.050	40	300	0.011
19	21.70	2	0.000	40	150	0.000
20	21.86	3	0.040	40	20	0.0086
21	22.50	3	0.0004	40	200	0.00008
22	22.99	3	1.03	40	290	0.21
23	23.67	2	0.82	40	450	0,17
24	24.03	3	1.20	40	70	0.24
25	24.61	2	0.077	40	100	0.016
26	25.64	2	0.006	40	50	0.0012
2/	26.33	3	3.9	40	270	0.77
28	27.27	3	0.004	40	/50	0.0008
29	28.70	2	0.1	40	/50	1.15
30	29.59	2	0.59	40	50	0.108
32 31	30.05	3	0.00	40	20	0.000
22	30.91	ר י	2.3	40	260	0.42
33	33.21	2	0.22	40	150	0.039
24	33.00	2	0.31	40	100	0.054
32	34.13	2	0.10	40	300	0.01/
סג 7 כ	34.00	4	2.4	40	900	0.41
3/	30.1	2	0.24	40	500	0.041
30	30.03	4	0.00	40	900	0.000
37	3/.3/	5	0.20	40	500	0.032
40	38.1	د	U.48	40	230	0.077

In the multilevel analyses of Simpson et al. [28,29], Moore et al. [27], James [12] it was assumed that there are one neutron and two fission channels and a large number of radiative capture channels. The analyses show two non-interfering groups of resonances; members of each group interfere among themselves. We identify the group of wide  $\Gamma_f$  with J = 2, and that of narrow  $\Gamma_f$  with J = 3 levels.

r	E <sub>r</sub> (eV)	J	Γ <sub>n</sub> (meV)	Γ <sub>γ</sub> (meV)	Γ <sub>f</sub> (meV)	Γ <sup>o</sup> (meV)
41	38.4	3	0.105	40	10	0.017
42	38.5	2	0.087	40	400	0.014
43	39.2	3	1.20	40	160	0.20
44	39.9	3	1.5	40	50	0.24
45	40.3	3	0.005	40	50	0.0008
46	40.83	2	2.9	40	1200	0.46
47	42.66	3	0.30	40	250	0.045
48	43.3	3	0.19	40	50	0.028
49	43.85	3	0.00	40	20	0.000
50	46.5	2	2.2	40	280	0.31
51	48.1	3	6.2	40	480	0.90
52	50.3	2	0.77	40	300	0.106
53	50.9	2	0.043	40	300	0.006
54	51.9	2	0.043	40	50	0.006
55	52.6	2	0.007	40	200	0.001
56	53.4	3	0.00	40	300	0.000
57	58.02	2	2.90	40	700	0.38
58	59.18	2	2.69	40	550	0.35
59	60.26	3	4.81	40	160	0.62
60	61.30	2	0.094	40	50	0.012
61	62.08	2.	8.12	40	650	1.03

The " $\Gamma_n^0$ " values of Simpson et al. [28] are thought to be closer to values for  $2g_J\Gamma_n^0$ , as in Moore et al. [27], and are treated as much. In Table IV,  $\Gamma_n^0$  is calculated from Simpson's data using the appropriate statistical factor  $g_I$ .

3.2.2. Average resonance parameters below 1 keV (Table V)

The nuclear momentum and parity of <sup>241</sup>Pu is  $I\pi = \frac{5}{2} +$ ; s-wave neutrons lead to states with J = 2,3 while p-wave neutrons lead to J = 1, 2, 3, 4. The recommended  $\langle \Gamma_{\gamma} \rangle$  is taken to be 40 meV [27-29].

The level spacings for the two s-wave sequences were computed and added "in parallel" to obtain the observed average spacing. If we adopt  $\langle D \rangle_{obs}$  and the expression for  $\langle D \rangle_{j}^{\ell}$  as in <sup>240</sup>Pu, we get the following "theoretical" values:  $\langle D \rangle_{j=1}^{\ell} = 2.98 \text{ eV}; \langle D \rangle_{j=2}^{\ell} = 2.04 \text{ eV}; \langle D \rangle_{j=3}^{\ell} = 1.76 \text{ eV} = \langle D \rangle_{j=4}^{\ell}$ : The "experimental" values, which we obtain for  $\ell = 0$ , are:  $\langle D \rangle_{j=2}^{\ell} = 2.33 \pm 0.26 \text{ eV}$  (19 resonances); and  $\langle D \rangle_{j=3}^{\ell} = 1.59 \pm 0.14 \text{ eV}$  (calculated from 31 resonances). For J = 2,3 we recommend the experimental values.

 $\xi_{l=0}^{J}$  was first computed for the J = 2,3 sequences separately, but since the difference between the two quantities obtained was smaller than the errors, the statistical average was adopted instead.

$$\xi_{\ell=0}^{J=2,3} \equiv \xi_0 = (1.30 \pm 0.75) \times 10^{-4}$$

$\langle D \rangle_{obs}$ = 0.945±0.093 eV	
$\langle D \rangle_{J=2}^{\ell=0} = 2.33 \pm 0.26 \text{ eV}$	
$\langle D \rangle_{J=3}^{\ell=0}$ = 1.59±0.14 eV	
$\langle D \rangle_{J=1}^{\ell=1}$ = 2.98 eV	
$\langle D \rangle_{J=2}^{l=1}$ = 2.33 eV	
$\langle \mathbf{D} \rangle_{I=3}^{\ell=1} = 1.59 \text{ eV}$	
$\langle D \rangle_{i=1}^{\ell=1} = 1.76 \text{ eV}$	
$\langle \Gamma^{0} \rangle^{k=0} = 0.342 \pm 0.109 \text{ meV}$	
$(-n)_{I=2}$	
$\langle \Gamma_n^+ \rangle_{J=3}^{L=0} = 0.188 \pm 0.047 \text{ meV}$	
$\langle \Gamma_n^0 \rangle_{J=1}^{\ell=1} = 0.745 \text{ meV}$	
$\langle \Gamma_n^0 \rangle_{J=2}^{t=1} = 0.582 \text{ meV}$	
$\langle \Gamma_n^0 \rangle_{J=3}^{\ell=1} = 0.398 \text{ meV}$	
$\langle \Gamma_n^0 \rangle_{J=4}^{\ell=1}$ 0.440 meV	
$\xi_{\ell=0}^{J=2,3} = \xi_0 = (1.30 \pm 0.75) \times 10^{-4}$	
$\xi_{\ell=1}^{J=1, 2, 3, 4} = \xi_1 = 2.5 \times 10^{-4}$	
$\langle \Gamma_{\rm f} \rangle_{\rm J=2}^{l=0}$ = .443±120 meV	
$\langle \Gamma_{\rm f} \rangle_{I=3}^{\ell=0}$ = 171±43 meV	
$\langle \Gamma_{\rm f} \rangle_{\rm J=2}^{\ell=1}$ = 443 meV	
$\langle \Gamma_{\rm f} \rangle_{\rm J=3}^{\ell=1}$ = 171 meV	
$\langle \Gamma_{\rm f} \rangle_{\rm J=1, 4}^{\ell=1}$ = 284 meV	
$\langle \Gamma_{\gamma} \rangle_{J=1,2,3,4}^{t=0,1} = 40 \text{ meV}$	
$\nu_{\rm n}$ = 1 for ( $\ell$ , J) = (0, 2), (0, 3), (1, 1), (1, 4)	1)
$\nu_{\rm n}$ = 2 for (ℓ, J) = (1, 2), (1, 3)	
$\nu_f = 1;  \nu_\gamma = \infty$	
$\sigma_{\rm pot}$ = 10.3 barns	

 
 TABLE V.
 Pu-241 AVERAGE RESONANCE PARAMETERS
 BELOW 1 keV
This value may be compared with that of Craig et al. [8], namely  $\xi_0 = (1.9 \pm 0.8) \times 10^{-4}$ . The value of  $\xi_{\ell=1}^J$  was taken as for <sup>239</sup>Pu [2] and assumed to be independent of J.

The  $\langle \Gamma_f \rangle_J^{\ell=0}$  were computed from Table IV. No information is available on p-wave fission; for the sake of computation we took  $\langle \Gamma_f \rangle_J^{\ell}$  to be independent of  $\ell$ . For the J = 1, 4, series we took the statistical average of the s-wave fission widths.

The capture width  $\langle \Gamma_{\gamma} \rangle$  was assumed to be independent of J and  $\ell$ . The potential scattering cross-section was taken to be 10.3 b, as for <sup>239</sup>Pu [2].

Statistical distributions.  $\langle D \rangle$  is considered to follow a Wigner distribution.  $\Gamma_n^0$  is assumed to follow a X<sup>2</sup>-distribution, with the number of degrees of freedom  $\nu_n$  equal to the number of neutron channels:  $\nu_n = 1$  for ( $\ell = 0$ , J = 2,3) and ( $\ell = 1$ , J = 1,4);  $\nu_n = 2$  for ( $\ell = 1$ , J = 2,3). The distribution of  $\Gamma_\gamma$  is assumed to be a delta function. Simpson et al.[28] found that for the two s-wave sequences  $\Gamma_f$  has a X<sup>2</sup>-distribution with  $\nu_f = 1$ .

### 3.3. Plutonium-242

# 3.3.1. Resonance parameters: values for neutronics calculations (Table VI)

Most of the information available comes from two recent experiments. Auchampaugh et al. [18] made transmission measurements below 400 eV

TABLE VI.	Pu-242 RESONANCE PARAMETERS	: VALUES
FOR NEUTR	ONICS CALCULATIONS	
1		

 $\ell = 0$ ,  $J = \frac{1}{2}$  for all resonances

r	E <sub>r</sub> (eV) r	Γ <sub>n</sub> (meV)	Γ (meV) γ	$\Gamma_n^0$ (meV)
1	2.64	1.92	25.5	1.19
2	14.60	0.061	30	0.016
· 3	22.48	0.28	30	0.060
4	40.96	0.48	30	0.075
5	53.7	52	18	7.1
6	68,2	3.0	30	0.36
7	89.1	0.80	30	0.085
8	106.0	0.6	30	0.06
9	108.0	14.8	30	1.42
10	132.7	5.9	30	0.51
11	151.1	17.7	30	1.44
12	166	1.0	30	0.08
13	205.6	31.0	30	2.2
14	217.4	4.6	30	0.31
15	235.4	7.8	30	0.51
16	276	10.1	30	0.61
17	311	11.0	30	0.63
18	324	229	30	12.7
19	336	94.5	30	5.2
20	388	47.3	30	2.40

with a resolution of 10-90 nsec/m using 97-99%  $^{242}$ Pu targets. The data were area and shape analysed. Pattenden [30] made a similar measurement below 850 eV (area analysis below 320 eV) with a best resolution of 15 nsec/m using 91%  $^{242}$ Pu targets. For measurements between 1961 and 1958, see BNL-325 (1965).

The only value for the fission width is  $\Gamma_{\rm f}$  (2.64 eV) < 0.02 meV (BNL-325 (1965)). We took  $\Gamma_{\rm f}$  = 0.0 meV throughout. For most of the resonances it is assumed that  $\Gamma_{\rm v}$  = 30 meV.

3.3.2. Average resonance parameters below 1 keV (Table VII)

For <sup>242</sup> Pu (I $\pi$  = 0 +) the J-sequences are: s-wave, J =  $\frac{1}{2}$ ; p-wave, J =  $\frac{1}{2}$ ,  $\frac{3}{2}$ . For  $\langle \Gamma_{\gamma} \rangle$  we took the value assumed by the experimenters, i.e. 30 meV. The observed level spacing is  $\langle D \rangle_{obs}$  = 18.5 ± 2.0 eV (calculated from 19 resonances). Pattenden [30] gives  $\langle D \rangle_{obs}$  = 18.1 ± 4.5 eV. We assumed the same expression for  $\langle D \rangle_{f}^{f}$  as in the case of  $^{240,241}$ Pu. We

TABLE VII. Pu-242 AVERAGE RESONANCE PARAMETERS BELOW 1 keV

 $\langle D \rangle_{J=\frac{1}{2}}^{k=0,1} = \langle D \rangle_{obs} = 18.5 \pm 2.0 \text{ eV}$   $\langle D \rangle_{J=\frac{1}{2}}^{k=1} = 10.2 \text{ eV}$   $\langle \Gamma_n^0 \rangle_{J=\frac{1}{2}}^{k=1} = 1.85 \pm 0.60 \text{ meV}$   $\langle \Gamma_n^0 \rangle_{J=\frac{1}{2}}^{k=1} = 4.62 \text{ meV}$   $\langle \Gamma_n^0 \rangle_{J=\frac{3}{2}}^{k=1} = 2.55 \text{ meV}$   $j = \frac{1}{2} \\ \xi_{k=0} = \xi_0 = (1.00 \pm 0.42) \times 10^{-4}$   $\xi_{k=1}^{J=\frac{1}{2},\frac{3}{2}} = \xi_1 = 2.5 \times 10^{-4}$   $\langle \Gamma_f \rangle_{J=\frac{1}{2},\frac{3}{2}}^{k=0,1} = 0.0 \text{ meV}$   $\langle \Gamma_f \rangle_{J=\frac{1}{2},\frac{3}{2}}^{k=0,1} = 30 \text{ meV}$   $\nu_n = 1; \quad \nu_f = 1; \quad \nu_\gamma = \infty$   $\sigma_{pot} = 10.6 \text{ barns}$ 

	eV	barns			
Pu isotope	E	<ح > ۲	<sup>&lt;σ</sup> f <sup>&gt;</sup>	<σ <sub>T</sub> >	
240	600	7.1 <u>+</u> 0.5	0.16	33.9 ± 1.0	
	1000	4.6 <u>+</u> 0.3	0.16	$28.5 \pm 0.8$	
241	60	22.1	43.4	77.7	
	100	$16.1 \pm 1.3$	33.5 <u>+</u> 0.7	62.0 <u>+</u> 1.8	
	300	9.14	20.6	42.1	
	1000	4.5 <u>+</u> 0.4	10.5 <u>+</u> 0.6	26.9 <u>+</u> 0.5	
242	350	7.17 + 0.5	0.01	32.3 + 1.0	
	1000	$3.18 \pm 0.22$	0.01	$23.2 \pm 0.7$	

TABLE VIII. AVERAGE CROSS-SECTIONS BELOW 1 keV

 $\langle \sigma_n \rangle = \langle \sigma_n \rangle - \langle \sigma_\gamma \rangle - \langle \sigma_f \rangle$ 

get  $\xi_0 = (1.00 \pm 0.42) \times 10^{-4}$ ; Auchampaugh et al. [18] obtained  $\xi_0 = (0.95 \pm 0.40) \times 10^{-4}$ . For  $\langle \Gamma_f \rangle$  we took  $\langle \Gamma_f \rangle = 0.0$  meV.  $\xi_1$  is expected to change only little from one isotope to the next; we therefore take the same value as for plutonium-240, 241, namely  $2.5 \times 10^{-4}$ . The statistical distributions are taken to be the same as for <sup>240</sup>Pu.

## 3.4. Last resolved resonance up to.1 keV

By "last resolved resonance" we mean the last one for which a complete set of parameters is available (<sup>240</sup>Pu, 679 eV; <sup>241</sup>Pu, 62 eV; <sup>242</sup>Pu, 388 eV).

Average cross-sections were calculated with the computer code SELF [25]. These (recommended) results are displayed in Table VIII. The calculated points are to be joined smoothly. The quoted errors are of a statistical nature. For the fertile isotopes,  $\langle \sigma_{\rm T} \rangle$  and  $\langle \sigma_{\gamma} \rangle$  were also calculated using the fluctuation factors evaluated by Schmidt [2]; they agree with the recommended values within the stated errors. Throughout the range we assume the values  $\langle \sigma_{\rm f} \rangle$  (<sup>240</sup> Pu) = 0.16 b; and  $\langle \sigma_{\rm f} \rangle$  (<sup>242</sup> Pu) = 0.01 b (these are the 1 keV values).

#### 4. FAST NEUTRON ENERGY RANGE

We include under this heading data from 1 keV to 15 MeV. The following cross-sections are presented in graphical form: total  $(\sigma_T)$ , nonelastic  $(\sigma_x)$ , fission  $(\sigma_f)$ , capture  $(\sigma_\gamma)$ , inelastic  $(\sigma_n)$ , partial inelastic  $(\sigma_n^{E_i})$ ,  $(n, 2n) \equiv \sigma_{2n}$ , and  $(n, 3n) \equiv \sigma_{3n}$ .

<sup>"</sup> For the Pu isotopes in question, the only fast neutron measurements available are those of  $\sigma_f$ , and even these do not cover the whole range.

As a rule, we have taken the  $\sigma_f$  evaluated by Davey [31], who renormalized previous relative measurements using new absolute measurements of the  $^{235}$ U and  $^{239}$ Pu fission cross-sections; the new  $\sigma_f$  values are up to 10% lower than previously. Davey covered measurements up to July 1965.

As shown by the experimental data and the optical model,  $\sigma_{\rm T}$  is similar for nuclei of close mass numbers. It is also expected that there are similarities between fertile nuclei among themselves, and fissile nuclei among themselves. Thus above 0.1-0.2 MeV,  $\sigma_{\rm T}$  of  $^{240}$  Pu and  $^{242}$  Pu was taken as for  $^{238}$ U, and  $\sigma_{\rm T}$  of  $^{241}$  Pu as that of  $^{239}$  Pu. Below 0.1-0.2 MeV,  $\sigma_{\rm T}$  can be accounted for by s- and p-wave neutrons only; since the strength function  $\xi_0$  of the Pu isotopes differs by as much as 40% from the U isotopes,  $\sigma_{\rm T}$  in this region was calculated for each isotope from its average resonance parameters.

In the MeV range the compound elastic scattering becomes negligible; as a result, the non-elastic cross-section becomes identical with the absorption cross-section, which according to the optical model is similar for nuclei of close mass numbers. Therefore, in the MeV range  $\sigma_x$  is taken as equal to that of an appropriate <sup>239</sup> Pu or <sup>238</sup>U isotope (as for  $\sigma_T$ ).  $\sigma_\gamma$  was calculated in the range 1-800 keV for <sup>240</sup>Pu and <sup>242</sup>Pu, and in the range 1-50 keV for <sup>241</sup>Pu. Unless otherwise indicated, nuclear data for <sup>239</sup>Pu and <sup>238</sup>U were taken from the compilation of Schmidt [2].

The following quantities were also included in the evaluation: average parameters above 1 keV (which were used in the present calculations of average cross-sections); the average number of prompt neutrons per fission ( $\mathcal{D}$ ); the average scattering cosine in the lab system ( $\bar{\mu}_L$ ); the energy spectrum of secondary neutrons from fission ( $\chi(E^{\dagger})$ ).

### 4.1. Plutonium-240

4.1.1. Average parameters above 1 keV

The energy dependence of  $\langle D \rangle$  can be represented by  $(B+E)^2 \exp[-2\sqrt{a(B+E)}]$ , where B is the neutron binding energy and a is the level density parameter [32]. Values for a are very uncertain in the mass region of 240, but the value 38 MeV<sup>-1</sup> is preferred [33] at present since it has been shown to improve the calculated ZPR-III critical masses as compared with those obtained using the value 22 MeV<sup>-1</sup> in YOM [34]. The neutron separation energy for (<sup>240</sup>Pu + n) is B = 5.39 MeV [35].

The values of  $\xi_1$  (<sup>235</sup> U, <sup>238</sup> U, <sup>239</sup> Pu) were recently found to be around 2.5 × 10<sup>-4</sup>. Since  $\xi_1$  is an intrinsic property of the nucleus we do not expect very different  $\xi_1$ -values for the higher Pu isotopes.

Since the strength functions are, by definition, independent of energy (over small energy ranges) we may assume that in the Doppler range (i.e. up to some 50 keV) they are constant.

The energy variation of  $\langle \Gamma_f \rangle$  of <sup>240</sup>Pu was determined by finding the values which give a fit of  $\langle \sigma_f \rangle$ (E) above 1 keV, when the above recommended parameters are used and assuming a Wigner distribution for D and an X<sup>2</sup>-distribution for  $\Gamma_n$  and  $\Gamma_f(\nu_n = \nu_f = 1)$ . The code SELF[25] was used for the calculation. We summarize the recommended parameters:

 $\langle D \rangle (E) = \langle D \rangle_{obs} \times (B + E)^2 \exp[-2\sqrt{a(B + E)}]$ a = 38 MeV<sup>-1</sup> B = 5.39 MeV  $\xi_0 (E) = 1.37 \times 10^{-4}$   $\xi_1 (E) = 2.5 \times 10^{-4}$ E(keV) 1 10 25 50 100 200 500

 E(keV)
 1
 10
 25
 50
 100
 200
 500
 900

  $\langle \Gamma_f \rangle_{J=1/2, 3/2}^{\ell=0, 1}$  0.23
 2.0
 3.0
 5.0
 8.0
 20.0
 65.0
 630

4.1.2. Cross-sections

 $\sigma_{\rm T}$  (Figs 1,2).  $\sigma_{\rm T}$  was calculated from the average resonance parameters above 1 keV. In the lower energy region the high-valued  $\xi_0$  makes  $\sigma_{\rm T}$  higher for <sup>240</sup> Pu than for <sup>238</sup>Pu. Above 100 keV the two curves practically coincide. From 200 keV to 15 MeV we assume  $\sigma_{\rm T}$  (<sup>240</sup> Pu) =  $\sigma_{\rm T}$ (<sup>238</sup>U).  $\sigma_{\rm T}$  for <sup>238</sup>U was taken from the evaluation of Schmidt up to 10 MeV and from that of Parker [36] in the range 10-15 MeV.

 $\sigma_x$  (Figs 1,2). In the range 1 to 60 keV the non-elastic curve is the sum of  $\sigma_f$  and  $\sigma_\gamma$ . In the range 60 to 1000 keV,  $\sigma_n(E)$  was calculated with the recommended resonance parameters and statistical distributions. The



FIG.1. <sup>240</sup> Pu: Total, non-elastic, capture, inelastic cross-sections; keV range.



FIG.2. <sup>240</sup> Pu: Total, non-elastic, capture, inelastic, (n,2n), (n,3n) cross-sections; MeV range.



FIG.3. <sup>240</sup> Pu: Fission cross-sections; keV range.

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fall of  $\sigma_n(E)$  towards  $\sigma_{pot}$  is more rapid than in <sup>238</sup>U mainly because the compound elastic starts to decrease appreciably already at 500 keV, where the sharp rise in  $\Gamma_f(^{240}Pu)$  sets in, while in <sup>238</sup>U this situation does not occur below 900-1100 keV. Thus  $\sigma_x$ , obtained as the difference  $\sigma_T - \sigma_n$ , reaches the value of 3 barns already at 1 MeV, as compared with 2 MeV for <sup>238</sup>U. Between 1 and 2 MeV,  $\sigma_x(^{240}Pu)$  was interpolated smoothly. Above 2 MeV, where the compound elastic is negligible,  $\sigma_n(^{240}Pu)$  was assumed to be equal to that of <sup>238</sup>U; therefore,  $\sigma_x(^{240}Pu) = \sigma_x(^{238}U)$ .



FIG.4. <sup>240</sup> Pu: Fission cross-sections; MeV range.

 $\sigma_{\rm f}$  (Figs 3, 4). In the range 1-18 keV partial averages were calculated from the nuclear detonation data of Byers et al. [22]. In the range 15-170 keV our curve goes through the values of Gilboy et al. [37] and the 24-keV point of Perkin et al. [38]. The data of Gilboy et al. give the cross-section ratios <sup>240</sup> Pu: <sup>235</sup>U measured by the time-of-flight method; we normalized these values to the  $\sigma_{\rm f}$  (<sup>235</sup>U) recommended by Davey. From 250 keV to 4 MeV we followed the recommendations of Davey [31]. Between 4 and 15 MeV we adopted the experimental values of Henkel et al. [39] renormalized to  $\sigma_{\rm f}$  (<sup>235</sup>U) as given by Davey, and the values of Kazarinova et al. [40] and Nesterov and Smirenkin [41].

 $\sigma_{\gamma}$  (Figs 1, 2). In the range 1-1000 keV,  $\sigma_{\gamma}$  (E) was calculated with the code SELF [25] using the recommended average parameters and statistical distributions. From 1 to 400 keV,  $\sigma_{\gamma}$  is 1.8 - 2.1 times the  $\sigma_{\gamma}$  (<sup>238</sup>U); this can be compared with recommendations of  $\sigma_{\gamma}$  (<sup>240</sup>Pu) = 1.75 ×  $\sigma_{\gamma}$  (<sup>238</sup>U) based on a few measurements of  $\sigma_{\gamma}$  (<sup>240</sup>Pu) [34]. Between 1 and 2 MeV,  $\sigma_{\gamma}$  (<sup>240</sup>Pu) was interpolated smoothly. Above 2 MeV we took  $\sigma_{\gamma}$  (<sup>240</sup>Pu) =  $\sigma_{\gamma}$  (<sup>238</sup>U).

 $\sigma_{n'}$  (Figs 1,2). After the determination of the other partial cross-sections and  $\sigma_{x}$ , the inelastic cross-section was calculated as  $\sigma_{n'} = \sigma_{x} - (\sigma_{f} + \sigma_{\gamma} + \sigma_{2n} + \sigma_{3n})$ . The threshold for this reaction is 43 keV.



FIG. 5. 240 Pu: Partial inelastic cross-sections; keV range.

 $\sigma_{n'}^{EJ}$  (Fig. 5). There is no experimental information on the excitation curves for <sup>240</sup> Pu; the relative excitation curves ( $\sigma_n^{EJ} / \sigma_{n'}$ ) for <sup>238</sup>U were therefore used. The assumed level scheme for <sup>240</sup>Pu (Table IX(a)) was constructed as follows: the five lower excited levels of <sup>240</sup>Pu, which agree (approximately) in energy and parity with the corresponding levels of <sup>238</sup>U, were used as such; for higher energies, where there is no correspondence between the <sup>240</sup>Pu and <sup>238</sup>U levels, we took the <sup>238</sup>U levels. The <sup>240</sup>Pu levels were taken from Douglas [42].

 $\sigma_{2n}$ ,  $\sigma_{3n}$  (Fig.2). The  $\sigma_{2n}$  curve was constructed as follows. The  $\sigma_{n,M}$  cross-section for neutron emission was calculated as  $\sigma_{n,M} = \sigma_x - (\sigma_f + \sigma_\gamma)$ . The ratio  $(\sigma_{2n}/\sigma_{n,M})$  was then calculated from the theoretical work of Pearlstein [43];  $\sigma_{2n}$  is then given by  $\sigma_{n,M} \times (\sigma_{2n}/\sigma_{n,M})$ . The cross-section for the (n, 3n) process was calculated in a similar way. Threshold energies were taken from Douglas [42].

(a) Pu-240		(b) Pu-241		(c) I	(c) Pu-242	
E (keV)	Ιπ	E <sub>1</sub> (keV)	Īπ	E <sub>i</sub> (ke)	<b>ν) Ιπ</b>	
0	0+	0	<u>5</u> + 2		0 0+	
43	2+	33	$\frac{7}{2}$ +		45 2+	
142	4+	95	<u>9</u> + 2	(14	48) (4+)	
292	6+	(155)	$(\frac{11}{2}+)$	(3)	LO) (6+)	
597	1-	(229)	$(\frac{13}{2}+)$	(68	30) (1-)	
640	3-	* (313)	$(\frac{1}{2}-)$	(73	32) (3-)	
(838)	(5-)	(315)	( <u>15</u> +)	(8:	38) (5-)	
(939)	(0+)	(342)	, i	(93	39) (0+)	
<b>★</b> ∫ (968)	(2+)	(400)		<b>★</b> ∫ (96	ó8) (2+)	
(1006)	(0+)	(417)		<u>[(100</u>	)6) (0+)	

TABLE IX. ASSUMED LEVEL SCHEMES

The brackets indicate that the levels were taken from U-238 (a, c) or U-233 (b) \* Taken as one level.

### 4.1.3. Other nuclear data

 $\bar{\nu}$ . A linear fit was made to recent measurements of the average number of (prompt) neutrons per fission [44,45,46]. The recommended curve is  $\bar{\nu} = 3.00 + 0.101$  E(MeV).

 $\bar{\mu}_{\rm L}$ . The average elastic scattering cosine for <sup>240,241,242</sup> Pu is taken equal to  $\bar{\mu}_{\rm I}$  (<sup>238</sup>U).

 $X(E^{\dagger})$ . The energy spectrum of secondary neutrons from fission is taken to be equal to the Watt spectrum, as recommended for <sup>238</sup>U by Schmidt:

 $X(E') = 0.45270 \exp(-E'/0.965) \sinh(2.29 E')^{1/2}$ 

where E' is the secondary neutron energy.  $\langle E' \rangle = 1.98$  MeV.

### 4.2. Plutonium-241

4.2.1. Average parameters above 1 keV

See the discussion for  $^{240}$ Pu. The separation energy for ( $^{241}$ Pu + n) is B = 6.26 MeV [35]. The average fission widths are taken to be constant up to 50 keV:

 $\langle \Gamma_{\rm f} \rangle_{\rm J=2}^{\ell=0,1}$  = 443 meV;  $\langle \Gamma_{\rm f} \rangle_{\rm J=3}^{\ell=0,1}$  = 171 meV



FIG.6. <sup>241</sup>Pu: Total (keV and MeV range), and fission (keV range) cross-sections.



FIG. 7. <sup>241</sup> Pu: Non-elastic, capture, inelastic cross-sections; keV range.

### 4.2.2. Cross-sections

 $\sigma_{\rm T}({\rm Fig.\,6}).$  The curve for  $\sigma_{\rm T}({\rm E})$  in the range 1 to 200 keV was calculated from the recommended strength functions. Potential-resonance interference was included. Up to 10 keV,  $\sigma_{\rm T}(^{241}{\rm Pu})$  is somewhat higher than  $\sigma_{\rm T}(^{239}{\rm Pu})$ ,

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due to a higher  $\xi_0$  value (1.30 compared with 1.07). From 1 up to 200 keV the curve for <sup>241</sup> Pu runs along that for <sup>239</sup> Pu with only small deviations from it. Above 200 keV we assumed equal  $\sigma_T$  for <sup>239</sup>Pu and <sup>241</sup>Pu.  $\sigma_T(^{239}$ Pu) was taken from the evaluation of Schmidt up to 10 MeV and from Douglas [32] in the range 10-15 MeV.

 $\sigma_x$  (Figs 7,8). From 500 keV to 15 MeV we assumed  $\sigma_x$  (<sup>241</sup>Pu) =  $\sigma_x$  (<sup>239</sup>Pu). Below 500 keV,  $\sigma_f$  (<sup>241</sup>Pu) increases rapidly with decreasing energy and becomes greater than  $\sigma_x$  (<sup>239</sup>Pu); therefore in the range 1-500 keV we take for <sup>241</sup>Pu,  $\sigma_x = \sigma_f + \sigma_\gamma + \sigma_n$ , where  $\sigma_n$ , (<sup>241</sup>Pu) =  $\sigma_n$ , (<sup>239</sup>Pu).





 $\sigma_{\rm f}$  (Figs 6,8). In the range from 1 keV to 10 MeV we take the renormalized and evaluated data of Davey [31]. From 10 to 15 MeV we follow the measurements of Smith et al. [47].

 $\sigma_{\gamma}$  (Figs 7, 8). The recommended average parameters and distributions were used to calculate  $\sigma_{\gamma}(E)$  in the range 1 to 50 keV. The curve comes out above the one for <sup>239</sup> Pu but quite close to it, and above 50 keV it was joined smoothly to  $\sigma_{\gamma}(^{239}$ Pu). Equal capture cross-sections for <sup>239</sup> Pu and <sup>241</sup> Pu were assumed in the range 100 keV to 15 MeV. Such an assumption (for the total 1 keV - 15 MeV range, however) was previously adopted by YOM[34] and Douglas [48]. The curve for <sup>239</sup>Pu was taken from Schmidt [49] up to 10 MeV and extrapolated to 15 MeV.

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 $\sigma_n$  (Figs 7, 8). From the threshold at 50 up to 500 keV,  $\sigma_n$  (E) is essentially the curve for  $^{239}{\rm Pu}$  (adapted to the different threshold of  $^{241}{\rm Pu}$  (n, n')) as recommended by Douglas [48]. In the range 500 keV - 15 MeV, the inelastic cross-section was calculated from  $\sigma_n$  =  $\sigma_x$  - ( $\sigma_f + \sigma_\gamma + \sigma_{2n} + \sigma_{3n}$ ).



FIG. 9. <sup>241</sup> Pu: Partial inelastic cross-sections; keV range.

 $\sigma_n^{E_j}$  (Fig. 9). The same procedure was followed as for <sup>240</sup>Pu. Here <sup>241</sup>Pu was compared with <sup>233</sup>U. For the latter isotope we took the results of the Hausser-Feshbach analysis of Kalos et al. [50]. The <sup>241</sup>Pu levels were taken from Douglas. The assumed scheme for <sup>241</sup>Pu is presented in Table IX (b). We see that only two levels of <sup>241</sup>Pu can be identified with levels of <sup>233</sup>U.

 $\sigma_{2n},\,\sigma_{3n}({\rm Fig.}\,\beta)$  . These cross-sections were calculated in the same manner as for  $^{240}{\rm Pu}.$ 

### 4.2.3. Other nuclear data

 $\bar{\nu}$ . For  $\bar{\nu}$  (<sup>241</sup>Pu) we adopt the results of Condé et al. [51], which represent the only determination of the energy dependence of  $\bar{\nu}$  to date.

The least squares fit to their data is  $\bar{\nu} = 2.905 + 0.146$  E(MeV). At thermal energy there is a discrepancy between Condé's value and that recommended in BNL-325 (1965); but four of the more recent values listed there agree with Condé.

 $\bar{\mu}_1$ . The same as for <sup>240</sup>Pu.

 $\chi(E^{i})$ . This is taken to be equal to the Watt spectrum recommended for  $^{239}\mathrm{Pu}$  by Schmidt:

 $X(E^{\dagger}) = 0.48395 \exp(-E^{\dagger}) \sinh(2E^{\dagger})^{1/2}$ 

 $\langle E' \rangle = 2.00 \text{ MeV}$ 

4.3. Plutonium-242

4.3.1. Average parameters above 1 keV

See discussion for <sup>240</sup>Pu.

a = 38 MeV<sup>-1</sup> B = 5.03 MeV  $\xi_0(E) = 1.00 \times 10^{-4}$  $\xi_1(E) = 2.5 \times 10^{-4}$ 

4.3.2. Cross-sections

 $σ_T$  (Figs 10, 11). In the range 1 to 100 keV,  $σ_T$  was calculated from the recommended resonance parameters. Starting from 1 keV the  $σ_T(^{242}Pu)$ curve is somewhat higher than the  $σ_T(^{238}U)$  curve, but at 20 keV it already practically joins the latter. From 100 keV to 15 MeV we assumed  $σ_T(^{242}Pu)$ =  $σ_T(^{238}U)$ . The latter curve was taken from Schmidt up to 10 MeV and from Parker [36] in the range 10-15 MeV.

 $\sigma_x$  (Figs 10, 11). Up to 2 MeV,  $\sigma_x$  was calculated as the sum of  $\sigma_f$ ,  $\sigma_\gamma$  and  $\sigma_n$ . Between 3 and 15 MeV we assumed  $\sigma_x$  (<sup>242</sup> Pu) =  $\sigma_x$  (<sup>238</sup> U). Between 2 and 3 MeV the curve was interpolated.

 $\sigma_{\rm f}$  (Figs 10,11). The  $\sigma_{\rm f}$  curve in the range 1 keV to 1.7 MeV was taken from Davey [31]; it follows the (renormalized) measurements of Butler [52] in the range 140 keV to 1.7 MeV. Since this curve resembles the one for <sup>240</sup>Pu in this range, its continuation above 2 MeV and up to 15 MeV was taken equal to  $\sigma_{\rm f}$  (<sup>240</sup>Pu).

 $\sigma_{\gamma}$  (Figs 10, 11).  $\sigma_{\gamma}$  was calculated from the resonance parameters between 1 keV and 1 MeV. Below 100 keV it is about 30% higher than  $\sigma_{\gamma}$  (<sup>238</sup> U); this difference can be accounted for by the higher  $\xi_0$  and  $\langle \Gamma_{\gamma} \rangle$ . At about 1 MeV the two curves have approximately the same value. Between 1 and 15 MeV we assume  $\sigma_{\gamma}$  (<sup>242</sup> Pu) =  $\sigma_{\gamma}$ (<sup>238</sup>U).



FIG. 10. <sup>242</sup> Pu: Total, non-elastic, fission, capture, inelastic cross-sections; keV range.



FIG. 11. <sup>242</sup>Pu: Total, non-elastic, fission, capture, inelastic, (n,2n), (n,3n) cross-sections; MeV range.





 $\sigma_n \cdot (\text{Figs 10,11})$ . From threshold (45 keV) up to 2 MeV we assumed  $\sigma_n \cdot (^{242}\text{Pu}) = \sigma_n \cdot (^{238}\text{U})$ . Above 2 MeV, where  $\sigma_x$  is taken as that of  $^{238}\text{U}$ , we calculated  $\sigma_n \cdot = \sigma_x - (\sigma_f + \sigma_\gamma + \sigma_{2n} + \sigma_{3n})$ .

 $\sigma_n^{E_j}$  (Fig. 12). Only the first excited state has been observed. We followed the same procedure as with <sup>240</sup>Pu. The assumed level scheme is presented in Table IX (c).

 $\sigma_{2n}$ ,  $\sigma_{3n}$  (Fig. 11). These were constructed as for <sup>240</sup> Pu.

4.3.3. Other nuclear data

 $\bar{\nu}$ . No data are available, and we therefore take  $\bar{\nu}$  (<sup>242</sup> Pu) =  $\bar{\nu}$ (<sup>240</sup> Pu) = 3.00 + 0.101 E(MeV) neutrons/fission.

 $\bar{\mu}_{I}$ : X(E<sup>†</sup>). See <sup>240</sup> Pu.

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### DISCUSSION

W.B. LOEWENSTEIN: Were your recommended <sup>240</sup>Pu cross-sections checked against available integral data?

S. YIFTAH: No, they were not, but we do intend to carry out this check.

W.B. LOEWENSTEIN: Does the trend of your new data indicate an improvement on the results presented at the International Conference on Fast Critical Experiments and their Analysis, organized by Argonne National Laboratory from 10 to 13 October 1966?

S. YIFTAH: I am afraid I cannot comment on current trends before performing the calculations.

K.H. BECKURTS: Your paper states that the capture cross-section of <sup>240</sup>Pu in the energy range 1-400 keV comes out to be 1.8 to 2.1 times larger than the capture cross-section of <sup>238</sup>U; does this refer to the experimental <sup>238</sup>U capture cross-section or to a calculated one, and have you tested the SELF code calculations against experimental  $\sigma_r$  values?

M. SEGEV: As co-author of this paper I can tell you that as a check on the SELF code we compared the calculated  $\sigma_r$  curve for  $^{238}$ U with the curve recommended by J.J. Schmidt, from Karlsruhe, and found very good agreement.

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# CALCULATION OF TEMPERATURE-DEPENDENT EFFECTIVE NEUTRON CROSS-SECTIONS

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### Abstract

CALCULATION OF TEMPERATURE- DEPENDENT EFFECTIVE NEUTRON CROSS-SECTIONS. The reliability of the Doppler coefficient, which is of decisive importance for the safety of fast reactors, depends strongly on the error margins for the temperature derivatives of the effective neutron cross-sections used in multi-group perturbation theory. Because in the energy region relevant for the Doppler effect most resonances of the fertile and all resonances of the fissile material are still unresolved, the evaluation of cross-sections must start with statistical parameters extrapolated from resolved resonances and subject to large errors. This paper attempts to clarify quantitatively the interdependence between the most important statistical resonance parameters and the temperature derivatives of the effective cross-sections. After an outline of the method chosen for the evaluation of these temperature derivatives, some improvements are discussed. As the well-known J-function and its derivatives with respect to both variables play an important part in the calculation, efficient sub-routines for these functions are required. The tabulations published until now are not well suited for this purpose and therefore we have performed a new tabulation, interpolation and approximation with higher accuracy and less expense of storage capacity. Asymmetric interference scattering is included by generalizing the J-function and approximating the correction term. The contribution to the temperature derivatives resulting from overlapping between resonances of the same sequence depends on the choice of the resonance distribution law. We compared a  $\chi^2$  distribution with degree of freedom  $\nu = 10$  and the Wigner distribution.

The improved method is applied to a typical fast reactor with different sets of input data. The interdependence between statistical parameters and the resulting temperature derivatives is studied by calculations with 10% change in one parameter only. Given error margins for the statistical resonance parameters lead to the desired error estimates for the temperature derivatives of the effective cross-sections. Comparison with the equivalent changes in cross-sections for infinite dilution offers the possibility of checking these error margins.

### 1. Introduction

The calculation of a fast-reactor Doppler coefficient by multi-group perturbation theory requires the temperature derivatives of effective reaction cross-sections. Because the accuracy of the Doppler coefficient is of decisive importance for the safety of the reactor, the error margins of these temperature derivatives must be known.

In the energy region relevant for the Doppler effect most resonances of the fertile and all resonances of the fissile material are still unresolved. The evaluation of cross-sections must, therefore, start with statistical parameters, extrapolated from resolved resonances and subject

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to large errors. The aim of this work is to clarify quantitatively the interdependence between the most important statistical resonance parameters and the temperature derivatives of the effective cross-sections.

For the calculation of effective cross-sections several more or less accurate methods have been proposed  $\begin{bmatrix} 1 & 7 & - & 5 & 7 \end{bmatrix}$ . We chose the method of Froelich  $\begin{bmatrix} 5 & 7 \end{bmatrix}$ , because it seems to combine high accuracy and relative simplicity. After an outline of this method we discuss some improvements, concerning the J-function and the overlap correction. Then we apply this method to the reactor investigated by Froelich and calculate the relative changes in the temperature derivatives of the capture and fission cross-sections caused by 10% changes in some statistical parameters. The results are compared with the equivalent changes in the corresponding cross-sections for infinite dilution.

2. Outline of method used

In multi-channel one-level theory the resonances of a nuclide are divided into sequences specified by the quantum numbers 1 and J. In each sequence s the distance  $D = D_{kk'}$  between successive resonance energies  $E_k$  and  $E_k$ , is distributed about a mean level spacing  $\overline{D}_s$  (see chapter 3.2).  $\overline{D}_s$  is dependent on energy according to

$$\overline{D}_{s} = \overline{D}_{J}^{\circ} \cdot \left(1 + \frac{E}{E_{B}}\right)^{2} \cdot \exp\left(-2 \cdot \sqrt{\alpha \cdot E_{B}} \cdot \left(\sqrt{1 + \frac{E}{E_{B}}} - 1\right)\right)$$
(1)

where  $\mathbf{E}_{\mathrm{B}}$  is the binding energy and a is a constant. The neutron widths  $\overline{\Gamma_{n,S}}$  and the fission widths  $\overline{\Gamma_{f,S}}$  are distributed about average values  $\overline{\Gamma_{n,S}}$  and  $\overline{\Gamma_{f,S}}$ , respectively, according to  $\chi^2$ -distribution laws with degrees of freedom  $\mathcal{V}_n$  and  $\mathcal{V}_f$ . The energy dependence of  $\overline{\Gamma_{n,S}}$  is given by

$$\overline{\Gamma_{m,s}} = S_{l} \cdot \overline{D_{j}} \cdot \sqrt{E} \cdot v_{l}$$
<sup>(2)</sup>

with strength function  $S_1$  and penetration factor  $v_1$  ( $v_0 = 1$ ;  $\frac{v_1}{r_{f,S}} = R^2/(R^2 + \lambda^2)$ , where  $R^2 \approx 6_p/(4\pi)$  and  $\lambda^2 = \Lambda^2/(E)$ . (E) must be taken from graphs. The distribution of the

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 $\gamma$ -widths  $\Gamma_{\gamma,s}$  is neglected,  $\overline{\Gamma_{\gamma,s}}$  is assumed constant. The sequence parameters  $1, J, \overline{D}_J^0, S_1, \nu_n, \nu_f, \overline{\Gamma_{f,s}}, \overline{\Gamma_{\gamma,s}}$  together with  $E_B$ , a,  $\mathbb{R}^2$ ,  $\Lambda^2$  and the atomic number densities  $N_s$  are the input parameters for cross-section calculations in the statistical energy region.

We consider an energy group with width  $\triangle$  E so small that the average resonance parameters can be taken as constant in it. In the narrow resonance approximation the group average effective macroscopic cross-sections for resonant reaction (n,z) (z=  $\chi$ , f, c =  $\chi$  + f+n) of sequence s are given by

$$\left\langle \mu_{z,s} \right\rangle^{\text{eff}} = \left\langle \frac{\mu_{z,s}}{\mu_{t}} \right\rangle / \left\langle \frac{1}{\mu_{t}} \right\rangle$$
(3)

where the total cross-section  $\mathcal{M}_t$  is the sum of the constant total potential scattering cross-section  $\mathcal{M}_p$  and all resonant cross-sections  $\mu_{cs}$  dependent on energy and temperature.

$$\mu_{t} = \mu_{p} + \sum_{s} \mu_{c,s} \qquad (4)$$

Instead of calculating the temperature derivative of  $\langle \mu_{z,s} \rangle^{eff}$ Froelich calculates only the temperature derivative of a fictitious cross-section defined by

$$\frac{d}{dT} \left\langle \stackrel{\wedge}{\mu}_{z,s} \right\rangle = \frac{1}{\left\langle \frac{1}{\mu_t} \right\rangle} \cdot \frac{d}{dT} \left\langle \frac{\mu_{z,s}}{\mu_t} \right\rangle$$
(5)

Froelich and Ott <u>6</u>7 have shown that this derivative gives in first-order perturbation theory the same result for the overall Doppler coefficient of a fast reactor as the derivative of eq. (3), if leakage and changes in elastic removal can be neglected. Since the correct splitting of the net effect into contributions of the individual sequences normally is not of interest, we adopt this method.

Froelich considers a homogeneous reactor composed of  $^{238}$ U and  $^{239}$ Pu oxide fuel with non-resonant coolant and structure material. He takes into account all l = 0 and l = 1 sequences, 3 for  $^{238}$ U and 5 for  $^{239}$ Pu. As the resonances of the l = 0 sequence of  $^{238}$ U (s = 1) are much higher than all

other resonances in the relevant energy region, they are treated separately.

In the calculation of  $\left< \frac{\ell^{\mu} \xi, 1}{\ell^{\mu} t} \right>$  interference with other sequences is neglected, i.e.  $\sum_{s>4} \ell^{\mu} \epsilon_{s,s}$  in the denominator is replaced by its average value  $\sum_{s>4} \langle \mu_{\epsilon,s} \rangle$  giving

$$\left\langle \frac{\mathcal{M}_{\mathcal{S},1}}{\mathcal{M}_{t}} \right\rangle = \left\langle \frac{\mathcal{M}_{\mathcal{S},1}}{\mathcal{M}_{p} + \mathcal{M}_{c,1} + \sum_{s>1} \mathcal{M}_{c,s}} \right\rangle \approx \left\langle \frac{\mathcal{M}_{\mathcal{S},1}}{\mathcal{M}_{p}^{\text{eff}} + \mathcal{M}_{c,1}} \right\rangle \tag{6}$$

with

$$\mu_{p}^{eff} = \mu_{p} + \sum_{s>1} \left\langle \mu_{c,s} \right\rangle \tag{7}$$

Similarly one gets

$$\left\langle \frac{1}{\mu_{t}} \right\rangle \approx \frac{1}{\mu_{p}^{eff}} \left\langle \frac{\mu_{p}^{eff}}{\mu_{p}^{eff} + \mu_{c,1}} \right\rangle = \frac{1}{\mu_{p}^{eff}} \left\langle \left(1 - \left\langle \frac{\mu_{c,1}}{\mu_{p}^{eff} + \mu_{c,1}} \right\rangle \right) \right\rangle$$
(8)

Because in sequence 1 the mean level spacing  $\overline{D}_1$  is large compared with the effective line widths, interaction between neighbouring resonances can be approximated by series expansion. Introducing the cross-sections for resonance k in  $\Delta E$ 

$$\mathcal{M}_{\mathcal{S},\mathcal{K}} = \mathcal{M}_{\mathcal{O}\mathcal{S},\mathcal{K}} \cdot \mathcal{\Psi}_{\mathcal{K}}$$

$$\mathcal{M}_{\mathcal{C},\mathcal{K}} = \mathcal{M}_{\mathcal{O}\mathcal{C},\mathcal{K}} \cdot (\mathcal{\Psi}_{\mathcal{K}} + \mathcal{\eta}_{\mathcal{K}} \cdot \chi_{\mathcal{K}})$$
(9)

where  $\psi$  and  $\chi$  are the usual symmetrical and asymmetrical line shape functions and  $\eta = \tan \phi_o$ , with phase shift  $\phi_o = \mathbb{R}/\mathcal{H}$  we get

$$\left\langle \frac{\mathcal{M}_{\mathbf{x},1}}{\mathcal{M}_{p}^{\text{eff}} + \mathcal{M}_{c,1}} \right\rangle = \left\langle \frac{\sum_{k} \mathcal{M}_{\mathbf{x},k}}{\mathcal{M}_{p}^{\text{eff}} + \mathcal{M}_{c,k} + \sum_{k' \neq k} \mathcal{M}_{c,k'}} \right\rangle \approx \\ \approx \sum_{k} \left\langle \left\langle \frac{\mathcal{M}_{\mathbf{x},k}}{\mathcal{M}_{p}^{\text{eff}} + \mathcal{M}_{c,k}} \right\rangle - \sum_{k' \neq k} \left\langle \frac{\mathcal{M}_{\mathbf{x},k} \cdot \mathcal{M}_{c,k'}}{\left(\mathcal{M}_{p}^{\text{eff}} + \mathcal{M}_{c,k}\right)^{2}} \right\rangle \right\rangle$$
(10)

Neglecting  $\gamma$  the first term can be expressed by the wellknown J-function (see chapter 3.1). The second term is simplified further. The denominator is replaced by its average value  $\langle \mu_t \rangle^2 = \left( \mu_p + \frac{\zeta}{s} \langle \mu_{c,s} \rangle \right)^2$ . Approximation of the  $\psi$ -function by

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a Gaussian according to Dresner  $\sqrt{77}$  finally leads to

$$\left\langle \frac{\mu_{z,1}}{\mu_{t}} \right\rangle \approx \frac{\overline{\Gamma_{z,1} \cdot J(\beta_{1}, \xi_{1})}}{\overline{D_{1}} \cdot \cos(2\phi_{0})} - \frac{\left\langle \mu_{z,1} \right\rangle \cdot \left\langle \mu_{c,1} \right\rangle}{\sqrt{2\pi} \cdot \left\langle \mu_{t} \right\rangle^{2}} \cdot \frac{\overline{D_{1}}}{\Delta} \cdot \varepsilon_{1}$$
(11)

The bar designates averaging over  $\Gamma_{n,1}$ .  $\beta_1 = \mu_p^{eff} / \mu_{oc,1}$ ,  $\xi_1 = \frac{\Gamma_1}{\Delta}$  with Doppler width  $\Delta$  and  $\varepsilon_1$  is an abbreviation for  $\varepsilon(\overline{D_1}/\Delta)$  with

$$\mathcal{E}\left(\frac{\overline{D}}{\Delta}\right) = 2 \cdot \int_{0}^{\infty} exp\left(-\frac{\overline{D}^{2}}{\Delta^{2}} \cdot \frac{x^{2}}{2}\right) \cdot \mathcal{Q}(x) \cdot dx \qquad (12)$$

where  $\Omega(\mathbf{x}) \cdot d\mathbf{x}$  is the number of resonances  $\mathbf{k'} \neq \mathbf{k}$  which lie in the interval dx about the relative distance  $\mathbf{x} = D/\overline{D}$  from resonance k (see chapter 3.2). With the similar abbreviation

$$e\left(\frac{\overline{D}}{\Delta}\right) = 2 \cdot \int_{0}^{\infty} \left(\frac{\overline{D}^{2}}{\Delta^{2}} - 1\right) \cdot exp\left(-\frac{\overline{D}^{2}}{\Delta^{2}} \cdot \frac{x^{2}}{2}\right) \cdot \Omega(x) \cdot dx \qquad (13)$$

the temperature derivative of eq.(11) may be written in the form

$$T \cdot \frac{d}{dT} \left\langle \frac{\mu_{\chi,1}}{\mu_{t}} \right\rangle = - \frac{\overline{\Gamma_{\chi,1} \cdot \Gamma_{1}} \cdot \frac{\partial}{\partial \xi} \int \left( \beta_{1}, \xi_{1} \right)}{2 \cdot \Delta \cdot \overline{D_{1}} \cdot \cos\left(2\phi_{0}\right)} - \frac{\left\langle \mu_{\chi,1} \right\rangle \cdot \left\langle \mu_{c,1} \right\rangle}{2 \cdot \sqrt{2\pi} \cdot \left\langle \mu_{t} \right\rangle^{2}} \cdot \frac{\overline{D_{1}}}{\Delta} \cdot e_{1} \quad (14)$$

The positive first term gives the main Doppler effect. It is somewhat reduced by the negative second overlap term.

The resonances of all sequences s > 1 are treated as a whole. Because they are low compared with  $\mu_p$  the following expansion is allowed  $(z = \chi_f)$ 

$$\sum_{s>1} \left\langle \frac{\mu_{z,s}}{\mu_{t}} \right\rangle = \sum_{s>1} \left\langle \mu_{z,s} \right\rangle \left[ \left( \mu_{z}^{\text{eff}} + \mu_{c,1} \right) \cdot \left( 1 + \frac{\mu_{p} - \mu_{z}^{\text{eff}} + \sum_{s>2} \mu_{c,s}}{\mu_{z}^{\text{eff}} + \mu_{c,1}} \right) \right] \right\rangle$$
(15)  
$$\approx \sum_{s>1} \left\langle \mu_{z,s} \right\rangle \cdot \left\langle \frac{1}{\mu_{z}^{\text{eff}} + \mu_{c,1}} \right\rangle - \left\langle \frac{\sum_{s>1} \mu_{z,s} \cdot \left( \mu_{p} - \mu_{z}^{\text{eff}} + \sum_{s>2} \mu_{c,s}}{\left( \mu_{z}^{\text{eff}} + \mu_{c,1} \right)^{2}} \right\rangle$$

The second term vanishes, if we choose

$$\begin{aligned}
\mu_{2}^{\text{eff}} &= \left[ \mu_{p} + \sum_{s>1} \sum_{s'>1} \left\langle \left[ \mu_{2,s} \cdot \mu_{c,s} \right] \right\rangle \right| \sum_{s>1} \left\langle \left[ \mu_{2,s} \right\rangle \right\rangle \\
&= \left[ \mu_{p}^{\text{eff}} + \sum_{s>1} \left( \left\langle \left[ \mu_{2,s} \cdot \mu_{c,s} \right] \right\rangle - \left\langle \left[ \mu_{2,s} \right] \right\rangle \cdot \left\langle \mu_{c,s} \right\rangle \right] \right) \right| \sum_{s>1} \left\langle \left[ \mu_{2,s} \right\rangle \right\rangle \end{aligned}$$
(16)

The average product  $\langle \mu_{z,s}, \mu_{c,s} \rangle$  can be calculated similar to the overlap term in eq. (10), but in the sum k' = k is included. With

$$E_{z,s} = \frac{\mathcal{M}_{oz,s} \cdot \mathcal{M}_{oc,s} \cdot \Gamma_{s}^{2}}{\mathcal{M}_{oz,s} \cdot \Gamma_{s} \cdot \mathcal{M}_{oc,s} \cdot \Gamma_{s}}$$
(17)

the bars denoting averaging over  $\vec{l}_{n,5}$  and  $\vec{l}_{r,5}$  , we get

$$\mathcal{M}_{z}^{eff} = \mathcal{M}_{p}^{eff} + \frac{\sum_{s>1} \langle \mathcal{M}_{z,s} \rangle \cdot \langle \mathcal{M}_{c,s} \rangle \cdot \left[ \frac{\overline{\mathcal{D}}_{s}}{\Delta} \cdot \left( E_{z,s} + \varepsilon_{s} \right) - \sqrt{2\pi} \right]}{\sqrt{2\pi} \cdot \sum_{s>1} \langle \mathcal{M}_{z,s} \rangle}$$
(18)

and

 $\hat{k}$ 

$$T \cdot \frac{d \mu_{z}^{\text{eff}}}{dT} = - \frac{\sum_{s>1} \langle \mu_{z,s} \rangle \cdot \langle \mu_{c,s} \rangle \cdot \frac{D_{s}}{\Delta} \cdot (E_{z,s} - e_{s})}{2 \sqrt{2\pi} \cdot \sum_{s>1} \langle \mu_{z,s} \rangle}$$
(19)

Transforming eq. (15) similar to eq. (8) yields finally

$$T \cdot \frac{d}{dT} \sum_{s>1} \left\langle \frac{M_{\tilde{z},s}}{M_{\tilde{z}}} \right\rangle \approx \frac{\sum_{s>1} \left\langle M_{\tilde{z},s} \right\rangle}{M_{\tilde{z}}^{eff}} \cdot \left[ \left( 1 - \left( \frac{M_{c,1}}{M_{\tilde{z}}^{eff} + M_{c,1}} \right) \right) \left( -\frac{T}{M_{\tilde{s}}^{eff}} \cdot \frac{dM_{\tilde{z}}}{dT} \right) - T \cdot \frac{d}{dT} \left\langle \frac{M_{c,1}}{M_{\tilde{z}}^{eff} + M_{c,1}} \right\rangle \right] (20)$$

The first term is positive and corresponds to eq.(14). The second term is negative and takes into account the interaction between sequences s > 1 and sequence s = 1.

3. Improvements

3.1 The J-function

Evaluation of eqs. (14) and (20) requires the J-function and its derivatives with respect to  $\beta$  and  $\xi$ . 4-digittabulations of this function have been published by several authors. Dresner's table [7] is restricted to  $0.1 \le \xi \le 1$  and  $0 \le k \le 31$  with  $\beta = 2^k \cdot 10^{-5}$ . Extensions to smaller  $\xi$  by Adler et al.  $[8_7]$  and by Nather et al.  $[9_7]$  have errors that amount to about 1.5 % for  $\xi = 0.05$  and  $\beta \approx 0.1$ . This would cause large errors in the derivatives. The best and most extensive table with about 10 000 function values is that of Bell et al.  $[10_7]$ , but it has some drawbacks, too. For large  $\beta$  the  $\xi$ -dependence of J is so weak that the last digit will change by one unit only after many steps. Besides waste in storage capacity this fact causes jumps in  $\partial J/\partial \xi$  which may disturb comparison of results for only slightly varied input data. Another drawback is the spacing of the  $\xi$ -scale, which is linear in several sub-intervals. This causes excess information at the upper and interpolation errors at the lower end of each sub-interval.

For these reasons we performed a new tabulation correct to 6 digits with an approximately logarithmic spacing of the  $\xi$ -scale. In order to get as continuous and smoothly varying derivatives as possible, we use a 4 times 4 point interpolation routine. Thus we reach an accuracy in J better than  $2 \cdot 10^{-4}$  with only 24  $\xi$ -points between 0.005 and 1 and 32  $\beta$ -points between k = 1 and k = 16.5 that are in all 776 function values. For k > 16 we get the same accuracy with the following rational expression.

$$J\left(\beta,\xi\right) = \int_{0}^{\infty} \frac{\psi(x,\xi) \cdot dx}{\beta + \psi(x,\xi)} \approx \frac{\pi}{2 \cdot \beta} - \frac{\alpha_{1} \cdot \xi + \alpha_{2} \cdot \xi^{2}}{1 + \alpha_{3} \cdot \xi + \alpha_{4} \cdot \xi^{2}}$$

with  $\mathbf{a}_{1} = 0.0013129 \cdot \beta^{-1} + 0.9818294 \cdot \beta^{-2}$  (21)  $\mathbf{a}_{2} = 0.35504947 \cdot \beta^{-2} + 0.17116913 \cdot \beta^{-3} - 0.023305 \cdot \beta^{-4}$   $\mathbf{a}_{3} = 1.1486697 + 0.8985628 \cdot \beta^{-1} - 0.0319557 \cdot \beta^{-2}$  $\mathbf{a}_{4} = \frac{2}{\pi} \cdot \alpha_{2} / (\beta^{-1} - \beta^{-1/2} \cdot (1+\beta)^{-1/2})$ 

Similar but simpler approximations are applied for k < 1.5and for  $\xi < 0.005$  or  $\xi > 1$  with  $1.5 \le k \le 16$ , because these regions are practically never used. Altogether this new approximation  $\sum 11_7$  gives higher accuracy in the J-function and its derivatives than any other method known to us and all this with drastically reduced expense of storage requirement compared with Bell's tabulation. The neglect of  $\eta$  in eq. (9) does not introduce serious error, but it always causes somewhat too large temperature derivatives, because the generalized J-function

$$J(\beta,\xi,\eta) = \int_{0}^{\infty} \frac{\psi(x,\xi) \cdot dx}{\beta + \psi(x,\xi) + \gamma \cdot \chi(x,\xi)}$$
(22)

with

$$\frac{\widetilde{n}}{2 \cdot \sqrt{\beta \cdot (4+\beta) - \frac{1}{4} n^2}} = \int \left(\beta, \infty, \gamma\right) < \int \left(\beta, \xi, \gamma\right) \leq \int \left(\beta, 0, \gamma\right) = \frac{\widetilde{n}}{2 \cdot \beta}$$

varies less with  $\xi$  than  $J(\beta, \xi, 0)$ . In order to be on the safe side without wasting machine time we made the following approximation

$$J(\beta,\xi,\gamma) \approx J(\beta,\xi,0) + \left[J(\beta,\infty,\gamma) - J(\beta,\infty,0)\right] \cdot f(\beta,\xi)$$
(23)

With a rational expression for  $f(\beta, \xi)$  an accuracy of few per cent can be reached, if  $\frac{4}{4} \cdot \eta^2 \ll \beta$ , which is valid for normal dilution. This correction decreases the total  $d\langle \hat{\mu}_{\beta} \rangle/d\tau$  for the reactor discussed in chapter 4 by somewhat more than 1% for energies between about 3 and 50 keV.

### 3.2. Resonance overlapping

The distribution function  $\Omega$  (x) used in eqs. (12) and (13) is given by the sum

$$\Omega(\mathbf{x}) = \sum_{n=1}^{\infty} P_n(\mathbf{x})$$
(24)

where  $P_n(x).dx$  is the probability that the n-th neighbour of resonance k lies in dx about the relative distance  $x = D/\overline{D}$  from  $E_k$  and  $P_n(x)$  obeys the recurrence relation

$$P_{n}(x) = \int_{0}^{x} P_{1}(t) \cdot P_{n-1}(x-t) \cdot dt \qquad (n>1) \qquad (25)$$

Froelich  $\int 5_7$  assumed a  $\chi^2$ -distribution with degree of freedom  $\nu = 10$  for P<sub>1</sub>(x). Hence he could describe  $\Omega$  (x) in

analytical form. Schmidt  $\int 127$ , however, showed that new experimental results are better represented by the Wigner-distribution

$$P_{1,w}(x) = \frac{\overline{n}}{2} \cdot x \cdot exp\left(-\frac{\overline{n}}{4} \cdot x^{2}\right)$$
(26)

Therefore we calculated numerically the corresponding  $\Omega_{W}(\mathbf{x})$ and the functions  $\mathcal{E}_{W}(\mathbf{y})$  and  $\mathbf{e}_{W}(\mathbf{y})$  defined by eqs. (12) and (13). The results are compared with Froelich's functions in fig. 1. As with the Wigner-distribution small x-values are more probable than with the  $\chi^{2}$ -distribution,  $\mathbf{e}_{W}(\mathbf{y})$  is considerably larger than  $\mathbf{e}_{10}(\mathbf{y})$  for  $\mathbf{y} \geq 5$ . That takes effect especially on the widely spaced resonances of sequence 1 which supply the main part of the Doppler effect. For the reactor discussed in the next chapter replacing of the  $\chi^{2}$ -distribution by the Wigner-distribution reduced  $d\langle\hat{\mu}_{X}\rangle/dT$ for energies below about 10 keV by some per cent and slightly increased it for higher energies.

### 4. Discussion of results

The reactor considered by Froelich  $\int 5_7$  was composed of 25 vol-%  $^{238}UO_2$ , 5 vol-%  $^{239}PuO_2$ , 40 vol-% Na and 30 vol-% structure material. Hence follow  $\mathcal{M}_p \approx 0.35 \text{ cm}^{-1}$  and the atomic number densities  $N_U = 0.005575 \text{ cm}^{-1} \cdot \text{barn}^{-1}$ ,  $N_{Pu} = 0.001125 \text{ cm}^{-1} \cdot \text{barn}^{-1}$  for an assumed fuel density of 10 g·cm<sup>-3</sup> (not given in  $\int 5_7$ ). The effective temperature was 900 °K.

In a first calculation we used the same statistical parameters as Froelich did. They are for  ${}^{238}\text{U:S}_{0}=\text{S}_{1}=10^{-4}(\text{eV})^{-1/2}$ ,  $\overline{D}_{1/2}^{0} = 18.5 \text{ eV}$ ,  $\overline{D}_{3/2}^{0} = 10.08 \text{ eV}$ ,  $\nu_{n} = 1$ ,  $\overline{\lambda} = 24.6 \text{ meV}$ ,  $\mathbb{R}^{2} = 0.8515 \text{ barn}$ ,  $\mathbf{E}_{B} = 4.7 \text{ MeV}$ ,  $\mathbf{a} = 22.43/\text{MeV}$ ,  $\Lambda^{2} = 2.0895 \text{ barn} \cdot \text{eV}$ ; and for  ${}^{239}\text{Pu}$  :  $\mathbf{S}_{0} = \mathbf{S}_{1} = 10^{-4} (\text{eV})^{-1/2}$ ,  $\mathbf{D}_{0}^{0} = 10.12 \text{ eV}$   $\overline{D}_{1}^{0} = 3.57 \text{ eV}$ ,  $\overline{D}_{2}^{0} = 2.4 \text{ eV}$ ,  $\nu_{n} = \nu_{f} = 1$ ,  $\overline{\lambda}$  independent of sequence taken from  $(\underline{13}, \overline{\lambda}, \overline{\lambda}) = 38.7 \text{ meV}$ ,  $\mathbb{R}^{2} = 0.8356 \text{ barn}$ ,  $\mathbb{E}_{B} = 6.4 \text{ MeV}$ ,  $\mathbf{a} = 22.43/\text{MeV}$ ,  $\Lambda^{2} = 2.0894 \text{ barn} \cdot \text{eV}$ .

In a second calculation we took the new parameters given with error margins by Schmidt  $\int 12_7$ . They are for  $^{238}$ U:  $S_0 = (0.9 \pm 0.1) \cdot 10^{-4} (eV)^{-1/2}$ ,  $S_1 = (2.5 \pm 0.5) \cdot 10^{-4} (eV)^{-1/2}$ ,



FIG.1. Functions  $\Omega_{10}(x)$ ,  $\epsilon_{10}(y)$ ,  $e_{10}(y)$  for  $\chi^2$ -distribution with degree of freedom  $\nu = 10$  and  $\Omega_w(x)$ ,  $\epsilon_w(y)$ ,  $e_w(y)$  for Wigner distribution.



FIG.2. Temperature derivatives of fictitious cross-sections versus energy:  $d < \hat{\mu}_{\gamma} > /dT|_F$  and  $d < \hat{\mu}_f > /dT|_F$  for Froelich's data;  $d < \hat{\mu}_{\gamma} > /dT|_S$  and  $d < \hat{\mu}_f > /dT|_S$  for Schmidt's data.

The results, shown in fig. 2, are not split into shares for U and Pu, because introduction of the fictitious crosssection  $\hat{\mu}_z$  and mixing up of U and Pu-effects in a unique



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 $\mathcal{M}_{\mathbf{Z}}^{\text{eff}}$  makes this partition insignificant. We only separated the  $\chi$ - and f-components, because they are rather independent of each other and often used separately in perturbation calculations. We see that the newer input data raise  $d \langle \mathcal{M}_{\mathbf{X}} \rangle / d\mathbf{T}$ appreciably for  $\mathbf{E} \geq 10^4$  eV and lower it by about 8 % for  $\mathbf{E} \approx 1 \text{ keV}$ .  $d \langle \hat{\mathcal{M}}_f \rangle / d\mathbf{T}$ , on the contrary, is practically unaffected despite large changes in  $\overline{\Gamma_f}$ ,  $S_1$  and  $\overline{D}_0$ , except at high energies where its absolute value is negligible. Because  $d \langle \hat{\mathcal{M}}_f \rangle / d\mathbf{T}$  is small compared with  $d \langle \hat{\mathcal{M}}_g \rangle / d\mathbf{T}$  for  $\mathbf{E} \geq 500 \text{ eV}$ , we will confine our further discussion to the latter function.

In order to find those statistical parameters which influence strongest  $d \langle \hat{\mu}_{g} \rangle / dT$ , we repeated the second calculation with one parameter raised by 10 % and all the others unchanged. The resulting changes in  $d \langle \hat{\mu}_{g} \rangle / dT$  are shown in fig. 3 for the most important parameters of <sup>238</sup>U: S<sub>0</sub>, S<sub>1</sub>,  $D_{1/2}^0$ ,  $\overline{D}_{3/2}^0$  and  $\overline{k_g}$ . We see that for low and intermediate energies the errors in  $\overline{D}_{1/2}^0$  and  $\overline{k_g}$  and for high energies the errors in  $\overline{k_g}$ , S<sub>1</sub> and  $\overline{D}_{3/2}^0$  are most serious. If all the error margins given in  $\int 12^{-7}$  were independent of one another, maximum error margins for  $d \langle \hat{\mu}_g \rangle / dT$  of about 20 % near 1 keV, 30 % near 10 keV and 40 % near 100 keV would result. Approximately half of this error is contributed by the 20 % uncertainty in  $\overline{k_g}$ . Because this error margin is concluded from only a few resolved resonances and because the probable value of  $\overline{k_g}$  was used in the analysis of the other resonances so that the strength function values S<sub>0</sub> and S<sub>1</sub> depend on it, we suppose that the total error margins might be safely reduced to 10 % near 1 keV, 15 % near 10 keV and 20 % near 100 keV.

If we compare with the corresponding relative changes in the microscopic capture cross-section of <sup>238</sup>U for infinite dilution  $\langle \tilde{\sigma}_{\delta}^{g} \rangle$  caused by changes of one parameter V (see fig. 4), we find qualitative and quantitative analogies. The maximum relative errors of  $\langle \tilde{\sigma}_{\delta}^{g} \rangle$  should, therefore, be comparable with those of  $d \langle \hat{\mu}_{\delta} \rangle / dT$ . This corresponds to some values recommended by Schmidt in <u>[12]</u>7, but it is inconsistent with some experimental results. More exact measurements of  $\langle \tilde{\sigma}_{\delta}^{g} \rangle$  for the region of unresolved resonances would help much in assuring the error margins given above.



FIG.4. Relative change in  $<\sigma_{\gamma}^8>$  of <sup>238</sup>U caused by relative change in parameter v versus energy.

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The curves in fig. 3 are certainly dependent on reactor composition and temperature but they will be good enough for error estimation of similar reactors. Uncertainty in  $\mathcal{M}_P$  may be taken into account by means of the dashed curve added for this purpose.

### 5. Conclusive remarks

We tried further improvements of the calculational method but did not yet include them into our standard program. For instance, we studied the Gaussian approximation to the  $\psi$ -function. It effects mainly on the statistical averages  $E_{z,s}$  of eq. (17), but because the error in  $f(\xi) = \int^{\infty} \psi^2(\xi, x) \cdot dx$  is less than 0.4 % for  $\xi \le 0.005$ , less than 1.6 % for  $\xi \le 0.02$  and less than 4 % for  $\xi \leq 0.05$ , the overestimation of d  $\langle \hat{\mu}_{\kappa} \rangle / dT$ caused by the low resonances of sequences s > 1 cannot be large. Nevertheless we will try to include this correction, because it can easily be done with a polynomial expansion of  $f(\xi)$ . Judging of the corresponding effect on the functions e(y) and  $\xi(y)$  is more difficult, but it seems to be small compared with that caused by the change from  $\chi^2$  to Wignerdistribution. The errors coming from narrow-resonance approximation were investigated recently in detail by Hwang [14\_7 and found to be negligible. Summing up, we can suppose that further efforts in improving the methods of calculation do not pay until the error margins of the statistical resonance parameters can be reduced drastically.

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### DISCUSSION

H.W. KÜSTERS: I should first like to point out that in the paper by Froelich (Ref. [5] of your paper) the temperature derivation of the denominator is in fact taken into account – albeit approximately and your method certainly represents an improvement in this respect.

Did you perform Doppler effect calculations and how do these compare with data obtained by the simpler method?

A. MÜLLER: In the paper to which I made reference (KFK 367) Froelich and co-workers neglected the temperature derivative of the denominator of the effective cross-section, corresponding to a flux change which vanishes in first-order perturbation theory when integrated over the reactor. Perhaps you had in mind some other internal work of Dr. Froelich.

So far our calculations have been confined to cross-sections and their temperature derivatives.

J.L. ROWLANDS: I should like to know from Dr. Müller whether he took into account the effect of statistical sampling in assessing the accuracy of Doppler coefficient calculations. I notice that in his paper (SM-101/22) Dr. Segev quotes a somewhat larger standard deviation of 100% or more, depending on the particular selection of resonances, for the Doppler change in <sup>239</sup>Pu cross-sections between 700 and 900 eV.

A. MÜLLER: The effect of statistical sampling was taken into account by numerical integration over all distribution functions of partial line widths and resonance distances using Gaussian quadrature formulas.

The difference between the standard deviations given by Dr. Segev and by myself may be explained as follows: Dr. Segev's data relate only to the  $^{239}$ Pu cross-sections between 700 and 900 eV, whereas my data are valid for a reactor composed of 25%  $^{238}$ UO<sub>2</sub> and only 5%  $^{239}$ PuO<sub>2</sub>. The influence of the  $^{238}$ U resonances (s-wave) far exceeds the effect of the  $^{239}$ Pu resonances in this energy region.

M. SEGEV: I would just point out that the 100% standard deviation in  $^{239}$ Pu cross-section change with temperature is quoted in my paper as a result obtained by Keller and Kier.

# THE GROUP CROSS-SECTION SET KFK-SNEAK Preparation and results \*

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### Abstract

THE GROUP CROSS-SECTION SET KFK-SNEAK: PREPARATION AND RESULTS. For the calculation of integral quantities, measured in the Fast Zero-Power Assembly SNEAK, a new group cross-section set was prepared directly from microscopic data. These data are based on the latest evaluation of cross-section and resonance parameters tabulated on the Karlsruhe Nuclear Data File KEDAK. Special attention is paid to the calculation of the elastic moderation.

This group set is partially organized in a manner similar to the Russian ABN-set. This means a tabulation of isotope-dependent group cross-sections for infinite dilution and energetic self-shielding factors, tabulated for the parameters  $\sigma_0$ , describing the dependence on the mixture, and for the temperature. The neutron-spectrum  $\Phi(E)$  is replaced by  $\Phi(E) = F(E)/\Sigma_t(E)$ , where F(E) is the collision density referred to below as 'macroscopic weighting-function' and  $\Sigma_t(E)$ , the total cross-section of the mixture. A macroscopic weighting-function has been obtained by calculations with the ABN-set, and the Karlsruhe KFK-group set, smoothing the step-function for the collision density.

The elastic scattering matrix is calculated from experimental cross-sections on KEDAK for all light- and medium-weight isotopes at the beginning of every diffusion calculation. The dependence on the mixture is taken into account exactly. For heavy-weight materials a similar representation is used.

Inelastic scattering and the transport cross-section are treated in a similar way as in the ABN-set. For the calculation of the group cross-sections and self-shielding factors a fully automated codesystem has been established.

During the preparation of the SNEAK-set the influence of the new material-dependent group crosssections on various integral reactor parameters was studied.

The basic material composition for the calculations is the core composition of SNEAK-3A-2, with an equivalent steam density of  $0.07 \text{ g/cm}^3$ , which is different from that of SNEAK-3A-1.

The procedure was as follows: starting with the ABN-set, the cross-sections of <sup>238</sup>U, O, Fe, Cr, C, Ni, Al, <sup>235</sup>U were successively replaced. For the ABN-set and then after each material substitution the following quantities were determined by fundamental mode calculations: the critical buckling  $B_c^2$ ,  $k_{\infty}$ , for  $B_c^2$  the neutron-flux,  $\triangle B_c^2$  against the preceding material replacement, the migration area, the conversion ratio, the Doppler coefficient and the neutron-generation time, for the critical buckling of the ABN-set ( $B_c^2$  ABN) the neutron-flux,  $\triangle k$  for coolant loss and the steam-density coefficient of reactivity.

To get a more detailed insight into the effect of microscopic data on neutron balance and the spectrum, the authors calculated in each case, for all core materials and for <sup>239</sup>Pu, <sup>240</sup>Pu, <sup>241</sup>Pu (both for  $B_c^2$  and  $B_c^2$  ABN), shielded and unshielded microscopic one-group constants by condensing with the corresponding 26 group spectrum.

Furthermore, the effect of the more accurate method in calculating the elastic removal crosssection on  $k_{eff}$ ,  $B_c^2$ , spectrum and reaction rates was studied.

A precise investigation of the influence of the change in group-cross-sections and a comparison with some integral experiments in SNEAK-3A-1 are given.

\* Work performed within the association in the field of fast reactor between the European Atomic Energy Community and Gesellschaft für Kernforschung m.b.H., Karlsruhe.

### 1. INTRODUCTION

The lack of accuracy in some essential cross-section types gives rise to considerable fluctuations in some important quantities used in the design of large fast reactors. As long as only concepts are considered and trends of main integral nuclear parameters are of interest, the theoretical methods and basic data used today are able to give satisfactory answers. But in calculating the nuclear properties of a fast reactor, already built or under construction, with the feedback on thermodynamic and economic properties, safety and long-time characteristics one obtains remarkable differences with different basic data recommended by various authors. Looking at integral reactor quantities shown in the intercomparisons at the Argonne Conferences in 1965 and 1966 the accuracy in the calculation of fast reactors is improving with time, but convergence of the results has not yet been reached.

In Karlsruhe the fast critical assembly SNEAK has been operating since December 1966. The theoretical analysis of the experiments clearly should be done on the basis of the latest and best cross-section data and method. Up to 1965 we calculated fast reactors with the Russian group constant set ABN from Bondarenko et al. [1].

In 1965 Schmidt revised his first cross-section evaluation [2], and on this basis we prepared new infinite dilute group constants, with a weighting spectrum of a typical 1000-MW(e) sodium-cooled reactor, for the most important materials used in fast breeders. The resonance self-shielding factors were taken from the Russian set with only slight modifications in the shielding of elastic moderation in certain groups. Preparation of this group constant set KFK 26-10 and results for large power reactors and critical assemblies are given in Ref. [3].

In 1966 a second revision of microscopic data has been published by Schmidt [4]. The recommended cross-sections and statistical parameters are available on the Karlsruhe Nuclear Data File KEDAK. From these data we prepared the group cross-section set KFK-SNEAK in 26 groups with resonance self-shielding factors calculated by ourselves. It should be noted that no adjustment to the special assembly has been made. The term 'SNEAK' in the group set arises from the fact that as a weighting spectrum for averaging the cross-sections a smoothed 26-group collision density spectrum has been used which was calculated with KFK 26-10 for the assembly SNEAK 3A-2. The hydrogen density used in this core is also typical for a large steam-cooled fast reactor (0.07 g/cm<sup>3</sup>). This is twice the hydrogen density of the assembly SNEAK-1, which has been analysed experimentally during the last months [5].

This paper will give a short survey of the methods used in calculating the group constants. A comparison of the main differences in the basic data is shown. During the preparation of the group constant set the influence of the new data on interesting integral quantities for SNEAK 3A-2 is studied. For the assembly SNEAK 3A-1 under experimental investigation now the essential results and a first conclusion about the quality of the set is given. In a further section some results of cal-
culations with the SNEAK-set for ZPRIII/48 and for large sodiumand steam-cooled reactors will be discussed.

# 2. MICROSCOPIC GROUP CONSTANTS

# 2.1. Theoretical aspects

The main calculational techniques are described in this section. In addition to the determination of the elastic removal group constants, the effective microscopic group cross-section of type z in group g for isotope k is calculated by

$${}^{k}\sigma_{z}^{g} = \frac{\int \frac{k}{\kappa}\sigma_{z}(E)}{\int \frac{k}{\sigma_{t}(E) + \sigma_{0}^{g}} F(E)dE}$$
$$\int \frac{\int \frac{F(E)}{\sigma_{t}(E) + \sigma_{0}^{g}} dE}{\int \frac{F(E)}{\kappa}\sigma_{t}(E) + \sigma_{0}^{g}} dE$$

The collision density F(E) is a slowly varying function with respect to energy and composition. In the broad structural resonances F(E) is proportional to the effective logarithmic energy decrement of the mixture. A correction can, therefore, be applied to F(E), but it is not yet included in the group set presented here.

Correctly the denominators in Eq. (1) should contain in NR approximation the total macroscopic cross-section of the mixture under consideration. For practical purposes it is convenient to replace the cross-section of the rest materials by an appropriate average in group g. Usually one takes the infinite dilute total group cross-sections in group g for this average, but it is very unrealistic to take the average total crosssection of <sup>238</sup>U with a resonance level spacing of 18 eV as a background for the shielding of <sup>235</sup>U or <sup>239</sup> Pu resonances with level spacings of roughly one order of magnitude less. Therefore, we defined  $k \sigma_{0}^{g} = (1/N_{k}) \sum_{m \neq k} N_{m}^{m} \sigma_{t}^{g}$ , with <sup>238</sup>  $\sigma_{t}^{g}$  replaced by the potential scattering cross-section.

The effective group cross-section is split into two parts:

$${}^{k}\sigma_{z}^{g} = {}^{k}\sigma_{z,\infty}^{g}, \quad {}^{k}f_{z}^{g} \quad (T,\sigma_{0}^{g})$$
<sup>(2)</sup>

 ${}^{k}\sigma_{2,...}^{g}$  is the group constant for infinite dilution for T=0 and T is the average temperature of the fuel. Both factors are tabulated on the Karlsruhe GROUCO file. This splitting procedure is very useful in the statistical resonance region, because there  ${}^{k}\sigma_{2,...}^{g}$  is then calculated by integration over pointwise tabulated microscopic cross-sections, while the self-shielding factor is determined by statistical resonance parameters. By this procedure the influence of uncertainties in statistical parameters is reduced.

(1)

The evaluation of the self-shielding factors is based on the singlelevel Breit-Wigner formula. Interference of potential and resonance scattering and resonance overlap is taken into account. The detailed description of the calculational technique will be given in the full documentation of the SNEAK set.

For the description of elastic moderation the  $\sigma_0$  concept has been abandoned; the dependence on the mixture is taken into account in NR approximation exactly in each reactor calculation. For high-energy anisotropic scattering in the CMS system the known experimental angular distributions are directly inserted and integrated over the degradation interval. For the essential energy region of the important resonances the experimental data provided are sufficient; they are continually being brought up to date. The automatic calculation of the macroscopic elastic removal group cross-section in every diffusion run necessitated a reorganization of the KEDAK file, which resulted in a new ERDAK file which is used in these calculations.

The transport cross-section is defined by

$$\sigma_{\rm tr}^{\rm g} = \sigma_{\rm t}^{\rm g} - \sum_{j \le g} \sigma^{j \to g} \cdot \mu^{j \to g}$$
(3)

where  $\sigma^{j \rightarrow g}$  is the scattering matrix (calculated with the appropriate weighting F(E)) and  $\mu^{j \rightarrow g}$  are the elements of the matrix for the average scattering cosine. Equation (3) is based on the assumption that the neutron current is only weakly dependent on energy, at least within the most important neighbouring groups to group g. In the SNEAK set up to now only hydrogen is treated by Eq. (3), because for isotopes with higher atomic weight than 10 the second term on the right-hand side of Eq. (3) can be approximated by  $\sigma_{g}^{g}$ .  $\mu_{g}^{g}$ .

The inelastic scattering matrix is calculated by taking the same transfer function as in ABN but normalizing the matrix elements to the re-evaluated total inelastic cross-section. The (n, 2n) processes are included in the inelastic matrix.

The various codes for calculating the group constants are combined in the code system MIGROS. The organization is given in Fig. 1. Two different methods can be used in calculating the elastic moderation: method A is based on the  $\sigma_0$  concept, method B calculates directly the shielded macroscopic elastic removal from the ERDAK file. The code system is fully automized, which is important especially for the determination of shielding factors in the different energy regions of resolved, statistical and strongly overlapping resonances.

# 2.2. Comparison of group constants

The SNEAK set contains new group constants, based on a re-evaluation by Schmidt, for the following elements or isotopes: H, C, O, Na, Al, Cr, Fe, Ni, <sup>235</sup>U, <sup>238</sup>U, <sup>239</sup>Pu. The cross-sections for other isotopes are, up to now, identical with those given in the ABN set. New group constants will be provided also for fission products [6] and the higher plutonium isotopes [7], but these data had not yet been included on the KEDAK and GROUCO files during the analysis discussed in this paper.

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As mentioned above, all new effective group cross-sections are weighted with the collision density F(E) which was obtained by smoothing the core averaged F(E) of a diffusion calculation with KFK 26-10 for SNEAK 3A-2 with the hydrogen density of 0.07 g/cm<sup>3</sup>. Only the main differences in the group constants of the SNEAK set compared to KFK 26-10 and the ABN set are shown here.



FIG.1. The code system MIGROS and the preparation of macroscopic effective group constants.

Figure 2 shows the fission and capture group cross-sections for  $^{235}$ U. SNEAK data for capture between 10 and 500 keV are smaller by 10-20% than the KFK data; the fission values, based on White's measurements, show the same behaviour, the deviations are, however, somewhat smaller. The low  $\sigma_c$  values result from lower  $\alpha$  data in this energy range.

Capture and fission group cross-sections for  $^{239}$ Pu are given in Fig. 3. Between 10 and 100 keV the fission data are taken from White's measurements (in Ref. [4] these values had not yet been recommended), and the low capture values result also from smaller  $\alpha$  data. The new data are smaller by 10 - 20% compared to the previous KFK data.

The capture cross-sections for Cr, Fe, and Ni are practically the same as in the KFK 26-10 set. The large differences in the region 10-100 keV, where the values used in Karlsruhe for  $\sigma_c$  (Fe) (Moxon measurements) are about a factor 2-3 times larger than the Russian constants should again be noted; for  $\sigma_c$  (Ni) there is a factor of 4, especially between 10-20 keV.

The Al capture group constants in the SNEAK set are larger than the ABN values in the essential energy region between 10 - 500 keV by 20% up to a factor of 2. We also have larger capture values in the low-energy region. The microscopic data for Al are taken from AEEW M445 with only slight modifications.

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The group constants for neutron capture in  $^{238}$  U are shown in Fig. 4, The SNEAK values differ in the energy region from 20 eV to 1 MeV by roughly 10-20% from the Russian data; compared to the first KFK data the new group constants are larger by about 10-15% between 1 and 20 keV,



FIG.2. Comparison of infinite dilute group cross-sections for capture and fission of <sup>235</sup>U.

smaller by about 20% in the regions 0.2 - 1 keV and 10 - 50 eV. The recommended capture data between 1 and 10 keV are calculated values from statistical theory and are just in between the measured data by Macklin et al. (1964) and Moxon et al. (1964). Very recently Pönitz and Menlove in Karlsruhe have measured the <sup>238</sup>U capture cross-section in the region between 10 and 500 keV [8]. These data are denoted in Fig. 4 by PM and are smaller than the SNEAK data by about 7-15%.

In Fig. 5 the self-shielding factors for  $\sigma_0 = 100$  barn and T = 900°K calculated with the recommended resonance parameters are compared with the equivalent ABN data. Between 10 eV and 1 keV the SNEAK values are larger by 10-30%.





To see the influence of the different methods in calculating the macroscopic elastic removal cross-section, the results for the SNEAK set are plotted in Fig.6. Method A is based on the  $\sigma_0$  concept; method B calculates the macroscopic group constant directly from microscopic data in each reactor calculation. Because the escape probability for a neutron scattered out of a group varies strongly within the degradation interval, and this interval for all isotopes except the very light ones is small against the group width and comparable to resonance widths of,

e.g., iron or oxygen, the removal cross-section of any isotope except the very light ones is strongly influenced by the resonance structure of the macroscopic total cross-section of the mixture. For the SNEAK assembly in two groups this influence is remarkable. In the group from



FIG.4. Comparison of infinite dilute group cross-sections for capture and fission of <sup>238</sup>U.

400 to 800 keV the oxygen resonance at 442 keV reduces the removal cross-section of all isotopes except hydrogen in method B, because the escape probability, weighted by the ascending wing of the total cross-section of oxygen in method B, is smaller than in method A, where only an average total cross-section is used. In the group from 21.5 to 46.5 keV the descending wing of the asymmetric resonance dip of iron at 25 keV causes a larger escape probability for all isotopes except hydrogen by method B than by method A. Therefore, the removal cross-section

is enlarged by method B. These effects can plainly be seen in the corresponding neutron spectra, shown in Fig. 11. The differences from the Russian removal cross-sections are quite large in the resonance region of structural materials, because besides the different weighting spectrum the infinite dilute value of the elastic scattering group constants as well as the corresponding shielding factor which enter the elastic removal cross-section are calculated for the total energy group and not for the degradation interval only.



FIG.5. Comparison of energetic self-shielding factors for capture ( $\sigma_n = 100$  barns, T = 900°K) of <sup>238</sup>U.



FIG. 6. Comparison of different procedures in calculating macroscopic elastic scattering matrix (see text).

Formula (3) yields a higher transport cross-section down to the keV region for hydrogen, and as a consequence the leakage of fast neutrons out of the system will be reduced. The previously used  $\sigma_{tr}^{H}$  data are calculated by  $\sigma_{tr}^{H} = \sigma_{s}^{H} (1 - \bar{\mu}) + \sigma_{a}^{H}$  where the average cosine of scattering is given by 2/3. Figure 7 shows the differences.



FIG. 7. Comparison of the transport cross-section of hydrogen.

# 3. RESULTS

# 3.1. Results obtained during preparation of KFK-SNEAK

The new constant set was prepared in a similar way as the first set KFK 26-10. As a basis we have taken the Russian ABN set and have replaced the group constants of the elements and isotopes mentioned above by new ones. After each replacement a fundamental mode diffusion calculation for the homogenized core using method A for elastic removal was performed to determine the influence of the new data on integral reactor quantities. The core composition is that of the assembly SNEAK 3A-2 with a hydrogen density similar to a larger steam-cooled fast reactor. The atomic number densities of the core materials are (the factor  $10^{24}$  has been omitted): N(Al) =  $1.289 \times 10^{-2}$ , N(C) =  $8.62 \times 10^{-4}$ , N(Cr) =  $3.42 \times 10^{-3}$ , N(Fe) =  $1.27 \times 10^{-2}$ , N(H) =  $1.64 \times 10^{-3}$ , N(Ni) =  $1.86 \times 10^{-3}$ , N(O) =  $1.446 \times 10^{-2}$ , N( $^{235}$ U) =  $2.03 \times 10^{-3}$ , N( $^{238}$ U) =  $8.108 \times 10^{-3}$ .

The fundamental mode caluclations were performed in two different ways: (1) achieving criticality by iteration of the buckling; (2) taking the critical buckling from a calculation with the ABN set and looking for the difference in criticality produced by the special replacement considered. The new group cross-sections for the various isotopes were replaced in the following sequence: (1)  $^{238}$ U, (2)  $^{16}$ O, (3) Fe, (4) Cr, (5)  $^{12}$ C, (6) Ni, (7) Al, (8) H, (9)  $^{235}$ U +  $^{239}$ Pu, (10)  $\sigma_p$  ( $^{238}$ U)+ $\sigma_{tr}^{H}$ , (11) Na.

In Figs 8-10 this sequence is numbered from 1 to 10; 1 corresponds to the replacement of  $^{238}$ U, 2 to the additional replacement of oxygen, replacement 10 introduces the potential scattering cross-section of  $^{238}$ U for  $\sigma_0$  determination and the  $\sigma_{tr}^{H}$  according to Eq. (3). With replacement 10 all materials which build up the core composition of the SNEAK assembly are inserted in the set. Na was replaced afterwards. On a separate tape we replaced the <sup>238</sup>U capture data recommended by Schmidt with the recent data of Ponitz and Menlove (PM data); all other materials are the same as in the SNEAK set.



FIG. 8. Changes in  $k_{eff}$  and  $k_{\infty}$  during preparation.

Figure 8 shows the behaviour of  $k_{eff}$  and  $k_{\infty}$  during the preparation procedure related to the corresponding values obtained with the ABN set, and Fig. 9 the total capture rate, <sup>238</sup>U-capture rate and the <sup>235</sup>U-fission rate. The integral quantities conversion ratio (CR), Doppler coefficient (DC) and neutron generation time ( $\ell$ ) are given in Fig.10. The corresponding results obtained with the KFK 26-10 set and with the PM capture data for <sup>238</sup>U are also included in all the figures. The results of Fig.8 are calculated with that buckling which produces  $k_{eff} = 1$  for the ABN set. All data in Figs 9 and 10 are taken from the corresponding critical system.

As a result of the replacement of  $^{238}$ U data in the ABN set there are less neutrons in the high energy region (larger inelastic scattering by about 15-10% in the region between 0.8 and 2.5 MeV) and more neutrons below 100 keV, mainly due to the changed capture data which are lower by about 15% between 50 and 400 keV compared to the ABN data. This spectral shift causes a small decrease in the  $^{238}$ U fission rate, an increase in  $^{238}$ U capture by 1%, (the average effective capture cross-section increases by about 2%), and as a consequence the k<sub>eff</sub> becomes smaller. The larger (1%) DC and CR are also due mainly to the spectral shift mentioned.

The oxygen replacement (2) results in a further softened spectrum, mainly because of the different weighting spectrum (F(E) = 1/E in ABN), giving higher capture rates, CR, DC and  $\ell$ .  $k_{\infty}$  is reduced because of larger absorption, but the leakage is even more reduced because at high energies more neutrons are slowed down, thus giving a smaller leakage component in the important high-energy region. For large reactors the  $k_{\infty}$  effect dominates the leakage effect.

The replacement of iron (3) merely results in an increase in the total capture rate giving less reactivity. This is caused by the higher capture cross-section (factor 2) in the new set.







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As the volume percentage of Cr, C, Ni is small in the SNEAK core, the effects of the replacements 4-6 are small.

Because of the better down scattering at high energies (weighting function) the Al replacement (7) gives less neutrons above 200 keV, more neutrons between 200 and 20 keV, and again less neutrons below 10 keV. Thus, the leakage is reduced, showing a larger k<sub>eff</sub> because k, is unchanged. The smaller number of neutrons below 10 keV is responsible for the remarkable decrease in DC. Hydrogen (case 8) scatters down more neutrons above 200 keV and less neutrons below because of the different weighting spectrum. This gives higher total capture rates and especially a remarkable increase in DC and  $\ell$ . The decrease in  $k_{\infty}$  is just compensated by the simultaneous decrease of the leakage rate. One should note, however, that in the ABN set we used Eq. (3) for calculating the transport cross-section, and in replacement 8 we had to take the usual  $\sigma_{tr}$  definition for organizational reasons. Thus, in this case the hydrogen is somewhat more transparent than in the other calculations, 1-7. The correction for  $\sigma_{tr}(H)$  is performed in replacement 10. The ninth replacement of <sup>235</sup>U gives by far the largest effects. The spectrum is softer because of a larger reduction of capture at lower than at higher energies. Thus, in particular the capture in <sup>238</sup>U becomes considerably larger by 10%, the fission and absorption become smaller because of the lower cross-sections despite the softer spectrum. In consequence,  $k_{\infty}$  becomes smaller (1%) and, as the leakage rate is also greater, k<sub>eff</sub> changes by about 1.7%. The larger capture in <sup>238</sup> U (spectrum effect) and the lower absorption in  $^{235}$ U (effect of crosssections) produce a 10% higher CR than in case 8. DC and l are also enlarged by roughly 10% (spectrum).

Case 10 contains the replacement of the hydrogen transport crosssection according to Eq. (3) (see also Fig. 7). Together with the new  $\sigma_{tr}^{H}$  the potential scattering cross-section of  $^{238}$ U  $\sigma_{p} = 10.6$  b has been introduced for the calculation of  $\sigma_{0}$  for the reasons mentioned in section 2.1. The leakage is somewhat decreased and overcompensates the decrease in  $k_{\infty}$ on account of the slightly softer spectrum.

Case 10 now is identical with the KFK-SNEAK group set. (The sodium replacement was done later because SNEAK 3A does not contain Na.) With this set calculations have been performed for the core composition SNEAK 3A-1. The results of these calculations are compared with experimental results in the paper of Stegemann et al. [5]. Before we draw the essential conclusions, we will discuss the effects of another cross-section replacement based on the recent measurements of the capture cross-section by Pönitz and Menlove (PM data). We obtained these data at a time when the calculations with the SNEAK set were almost complete and for consistency they were not included in the theoretical results of Ref. [5]. For the reference core composition discussed in this section (SNEAK assembly 3A-2) the lowered  $^{238}$ U capture results in an increase of k<sub>eff</sub> by 0.9% and the CR is lowered by 0.5%.

In Figs 11 and 12 the neutron spectra and adjoint spectra of the SNEAK 3A-2 core are shown for different sets and methods related to the values of the ABN set. In Fig. 11 the spectra calculated by method

	KFK 26-10	1	2	3	4	5	6	7	8	9	SN <b>E</b> AK	РМ
$\frac{\Delta k_{\rm L}}{(\Delta k_{\rm L})_{\rm ABN}}$	1.119	0 <b>.9</b> 325	0.8918	0.868	0.862	0.862	0.858	0.8198	0.8249	1.181	1.2008	1.180
$\frac{\frac{dk/k}{d\rho/\rho}}{\left(\frac{dk/k}{d\rho/\rho}\right)_{ABN}}$	1.076	0.9478	0.914	0.911	0.907	0.907	0.904	0.883	0.892	1.096	1.107	1.077

TABLE I.	CHANGES IN	LOSS OF	COOLANT	REACTIVITY	AND	REDUCED	STEAM	DENSITY	COEFFICIENT	
DURING PR	REPARATION									

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A and method B are given (for discussion see section 2.2). The sharp dip in the sodium resonance group 13 in the KFK 26-10 arises from the elastic removal cross-section (method A) of <sup>16</sup>O, which follows the Na resonance shape because of the weighting spectrum of a sodium-cooled reactor used in KFK 26-10. The changes in neutron importance can easily be understood by considering the changes in the <sup>238</sup> U capture and <sup>235</sup> U fission and capture group cross-sections given in Figs 2 and 4.



FIG.11. Comparison of neutron spectra.





Table I lists the changes in the loss of coolant reactivity  $\Delta k_L$  and the reduced steam density coefficient of reactivity  $(dk/k)/(d\rho/\rho)$ , both related to ABN values. The results can plainly be understood by

following the discussion given earlier. Again the largest differences are obtained in the  $^{235}$ U replacement in case 9. One should note the 20% decrease in  $\Delta k_L$  and 11% increase in the reduced steam density coefficient compared to ABN. The KFK 26-10 set gives results in between the ABN and SNEAK set, because a large part of microscopic data are similar to those incorporated in the SNEAK set. As we performed fundamental mode calculations using the critical ABN buckling, only the trends should be given precisely; clearly the absolute values must be obtained in 1- and 2-dimensional calculations.

# 3.2. Main results for the SNEAK 3A-1 assembly obtained with the KFK-SNEAK set

The detailed comparison of experimental and theoretical results obtained with the KFK-SNEAK set (method B) for the assembly 3A-1 is given in Ref. [5]. Here we will quote the main results to give an impression of the quality of data and methods used.

3.2.1. Criticality

Homogeneous two-dimensional diffusion theory, method B:	k hom eff	= 0.9895
$S_4$ -correction:	$\Delta k(S_4)$	= +0.0027
Heterogeneity correction: (calculated)	$\Delta k$ (het)	= + 0.0025
So we have	k <sup>het</sup> (SNEAK)	= 0,995
For comparison:	k <sub>eff</sub> <sup>hom</sup> (ABN)	= 1.019
	$k_{eff}^{hom}$ (KFK 26-10)	= 1.018

Accuracy of half a per cent in  $k_{eff}$  is the target of reactor calculations and this is achieved with the group set KFK-SNEAK. It should be noted again that no adjustments to measured reactor quantities have been made. Both KFK-26-10 and ABN give higher  $k_{eff}$  values by about 3%!

# 3.2.2. Spectrum and spectral indices

In Fig. 13 the spectra calculated with the SNEAK set (method A and method B) and with the KFK 25-10 set are compared with experimental results. Again we have satisfactory agreement between theory and experiment for method B and the SNEAK set. The less accurate method A gives 20% deviations in the most important groups for elastic scattering (oxygen and iron resonances). Inserting the PM data into the SNEAK set leaves the spectrum practically unchanged. The result can be improved by one iteration step for the collision density. From the spectrum, calculated by SNEAK-method B, a new collision density can be obtained which is used for calculation of new removal cross-section in another SNEAK method B calculation. This has not yet been done here, but the effects should not be large.

In Table II the most important fission and capture to fission ratios for the centre of SNEAK 3A-1 are compared with experiment. There is very good agreement in the <sup>238</sup>U capture to <sup>235</sup>U fission ratio, the other ratios do not fit as well to the experimental data. A check has been made in using the measured spectrum in the calculation of  $\sigma_f$  (<sup>238</sup>U)/ $\sigma_f$  (<sup>235</sup>U), giving no improvement. This effect is not yet understood and needs further investigation, especially with respect to the experimental technique used.



FIG. 13. Comparison of theoretical and experimental neutron spectra for assembly SNEAK 3A-1.

Table II also gives the corresponding ratios using the low <sup>238</sup>U capture data of Pönitz and Menlove. The drastic reduction in  $\sigma_c (^{238} U)/\sigma_f (^{235}U)$  is just within the experimental error line. With respect to criticality we obtain  $k_{eff}^{het}$  (PM) = 1.008 with the PM data, applying the same corrections for S<sub>4</sub> and heterogeneity given above. There is a strong indication that, because of normalization corrections, the fission and capture cross-sections of <sup>235</sup>U should be even smaller than the low White data included in the SNEAK set [10]. This would result in better agreement of the PM data with experiment. The influence of these renormalization effects on the important integral nuclear quantities is being investigated.

	Experiment	SNE AK SE T	KFK 26-10	ABN	ΡΜ σ <sub>c</sub> (U238)
$\frac{\sigma_{\rm f}(\rm U238)}{\sigma_{\rm f}(\rm U235)}$	0.0336± 3%	0.0301	0.0301	0.0316	0.0297
σ <sub>c</sub> (U238) σf(U235)	0.142± 0.008	0.143	0.129	0.127	0.136
σ <sub>f</sub> (Pu239) σ <sub>f</sub> (U235)	1.03 ± 3%	0.96	0.971	1.001	0.952

TABLE II. SPECTRAL INDICES IN THE CENTRE OF SNEAK 3A-1

## 3.2.3. Heterogeneity corrections

The SNEAK set gives far better agreement with the experimental results than both KFK 26-10 and the ABN set [5, 9]. The theory results in about 10% higher  $\Delta k^{het}$  than the measured results in the bunching experiments, while the ABN data are too small by a factor of two in  $\Delta k^{het}$ . Also the detailed cell fine structure gives much better results if the SNEAK set is used instead of the ABN or KFK 26-10 sets.

# 3.2.4. Void calculations

The sequence of void measurements in the central zone of SNEAK 3A-1 is fairly well represented by calculations with the SNEAK set [5]. Including heterogeneity corrections, there are deviations from experiment by about 5%. Results obtained with the ABN set are 30-40% too low (see also Ref. [9]).

# 3.2.5. Breeding ratio

Bearing in mind the theoretical and experimental results for the  $^{238}$ U capture to  $^{235}$ U fission ratio (Table II), the breeding ratio of SNEAK 3A-1 is also in much better agreement with experiment than results obtained with KFK 26-10 or ABN, see also Ref. [5].

# 3.3. Results for the ZPRIII/48 critical assembly

To check in particular the plutonium data incorporated in the SNEAK set, we performed a calculation for the ZPRIII/48 assembly and compared the results with those formerly obtained with the KFK 26-10 set [11]. This first calculation for the homogenized core was done with that core radius for which the KFK 26-10 set produces  $k_{eff}^{hom} = 1.0$ . With the

SNEAK set we obtained then  $k_{eff}^{hom}$  (SNEAK) = 0.963. The following corrections have to be made:

S4correction0.003Heterogeneity correction(as given in the ANL intercomparison) 0.014KFK 26-10 to experiment(as given in the ANL intercomparison) 0.011

Thus, on the basis of the ANL intercomparison we obtain with the SNEAK set  $% \left( {{{\rm{SNEAK}}} \right)$ 

$$k_{eff}^{het}$$
 (SNEAK) = 0.991

Some spectral indices are shown in Table III. Again, as in the case of SNEAK 3A-1, the agreement is not too good. A more precise calculation for the real geometry of ZPRIII/48 with redetermined heterogeneity corrections is under way.

# 3.4. Results for large fast power breeders

For completeness some calculations have been done with the SNEAK set for a steam-cooled large power reactor, the Karlsruhe D1 design [12]. A separate group set for the Karlsruhe prototype Na2 with sodium as a coolant was established with the appropriate core weighting spectrum and an additional blanket weighting spectrum. This set is named KFK-NAP [13]. The most important feature is an expected reduction in  $k_{eff}$  by about 2-3% for both types compared to results obtained with KFK 26-10. This means a difference in  $k_{eff}$  by about 6% compared to ABN calculations. For the steam-cooled reactor both coolant loss reactivity change  $\Delta k_L$  and the absolute value of the steam density coefficient are increased remarkably, changing the stability characteristics of this reactor for the worse. These investigations are not yet complete.

	Experiment	SNE AK SE T	KFK 26-10	ABN
$\frac{\sigma_{\rm f}(\rm U238)}{\sigma_{\rm f}(\rm U235)}$	0. 0307	0.0301	0.031	0.0328
σ <sub>c</sub> (U238) σ <sub>f</sub> (U235)	0.138	0.145	0.131	0.130
σ <sub>f</sub> (Pu239) σ <sub>f</sub> (U235)	0.976	0.91	0.938	0.958

TABLE III. SPECTRAL INDICES IN THE CENTRE OF ZPRIII/48

# 4. CONCLUSION

The comparison of the theoretical results, obtained with the new group constant set KFK-SNEAK, with the measured results for the fast critical assembly SNEAK 3A-1 show a remarkable and encouraging step forward in the field of fast reactor calculation. Compared to the previous KFK 26-10 set, the main reasons for the better agreement of experiment and theory are the low fission and capture data of <sup>235</sup>U included in the SNEAK set and an improved description of elastic moderation (method B), The remaining differences have to be analysed very carefully to get an idea of what has to be improved in microscopic cross-section evaluation and in the methods of establishing macroscopic self-shielding group constants. On the theoretical side, for instance, we are dealing with the problem of space-dependent resonance self-shielding (core-blanket interface), and in the near future we will have a code in consistent P1 approximation for 200 groups, so that the investigation of the spacedependent fine structure of the spectrum will, it is hoped, give further insight into the complex situation of fast reactor analysis.

# ACKNOWLEDGEMENT

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# DISCUSSION

M. SEGEV: In defining  $\sigma_{tr}$  you assumed a weak dependence of the current on the energy. Is this a valid assumption throughout the range of resonant cross-sections?

H.W. KÜSTERS: We redefined the transport cross-section for hydrogen only, and included the neutron current in a first iteration step, but this gave no further improvement. In order, inter alia, to eliminate simplifying assumptions, we decided to maintain a consistent  $P_1$  approach in multi-group (200) as well as in fewer-group calculations. In fact, it is not the current itself which is a weakly dependent function of the energy in the resonance region, but  $\Sigma_t^2$  J. Having once defined the corresponding group constants, one could clearly apply this to resonant cross-sections as well.

H.F. TROYANOV: Was a correction made for the form of the intragroup spectrum in the first KFK set of group constants? In the system of constants used by Bondarenko et al. this is allowed for by making successively more precise determinations of the spectrum.

H.W. KÜSTERS: As I mention in the Introduction, in calculating the group constants we used as a weighting spectrum a collision density spectrum with roughly 200 energy points, obtained by smoothing out the spectrum calculated for the SNEAK composition with our first KFK group set. This KFK set has a typical weighting spectrum for a 1000 MW(e) sodium-cooled reactor. We also iterated the SNEAK weighting spectrum sets but this gave no further improvement. With regard to elastic moderation, it was not the weighting spectrum alone which worried us, but rather the  $\sigma_0$  concept, which we therefore abandoned in favour of calculating the elastic slowing-down.

H.F. TROYANOV: Do you propose to make a detailed determination of the structure of the spectrum to obtain the cross-section of elastic transfer from one group to the next each time you do the reactor calculation?

H.W. KÜSTERS: In each reactor calculation we calculated the elastic removal cross-section for 1000 energy points directly form the KEDAK file using the fine-structure weighting  $1/\Sigma_t(E)$ , according to the narrow resonance approximation. The macroscopic F(E) weighting, which is a smoothly varying function of energy and composition, is changed only to allow for calculation of quite different systems, such as steam- or sodium-cooled reactors, or special fast facilities, e.g. STARK, SUAK or SNEAK.

# Closing remarks by Session Chairman

S. YIFTAH (Chairman): We have devoted this session to the discussion of various measured, calculated, evaluated and multigroup cross-sections, so before we end I should like to draw attention to cross-sections which we have not considered. I have in mind the potential importance of investigating the cross-sections and nuclear data of other less-common heavy nuclides, such as <sup>232</sup>U, the plutonium isotopes <sup>236</sup>Pu and <sup>238</sup>Pu, and certain isotopes of americium, curium and californium.

	US (tonnes/yr)	World (tonnes/yr)
Fission products	160	430
$^{236}$ U (in $^{235}$ U and $^{238}$ U)	22	60
<sup>237</sup> Np	2.3	6.3
<sup>238</sup> Pu	0.6	1.6
<sup>239</sup> Pu	40	110
<sup>240</sup> Pu	13	36
<sup>241</sup> Pu	6.7	18
<sup>241</sup> Am	0.9	2.5
<sup>242</sup> Pu	1.5	4.1
<sup>242</sup> Cm	0.03	0.08
<sup>243</sup> Am	0.6	1.7
<sup>244</sup> Cm	0.1	0.3

TABLE A. ANNUAL PRODUCTION OF ACTINIDE ISOTOPES

As regards the nuclear data requirements of the nuclear power industry, emphasis may well shift to new fields, at least in the United States of America. Whereas up till now efforts have been largely directed towards study of cross-sections for analysis and optimization of reactor design, safety and <u>initial</u> reactor operation, it seems that present trends look towards the nuclear data needs of power reactors operating for a relatively long time and producing large quantities of isotopic by-products from spent and recycled fuel. In this connection, problems of fuel burnup, prolonged exposure of sensitive components to neutron radiation and so on will be of prime importance. A full and detailed knowledge of the nuclear characteristics of all the nuclei present in a reactor at all times will enable it to be operated with greater reliability, at higher powers, to higher burn-ups, and with optimized isotope production.

According to an estimate by Snyder<sup>1</sup>, the annual production of the actinide isotopes in 1987 will be as shown in Table A.

Cross-sections of such isotopes as <sup>236</sup>Pu, half-life 2.85 yr, which is formed from <sup>238</sup>U after two (n, 2n) reactions or from <sup>235</sup>U after two (n,  $\gamma$ ) and one (n, 2n) reaction (in both cases via <sup>237</sup>Np), may assume great importance. <sup>236</sup>Pu is, in fact, at the head of a serious contamination chain, the <sup>236</sup>Pu  $\rightarrow$  <sup>232</sup>U  $\rightarrow$  <sup>228</sup>Th chain, in which all members following 1.91-yr <sup>228</sup>Th are short-lived isotopes, including many powerful gamma emitters.

Furthermore, the use of strongly alpha-active <sup>238</sup>Pu and <sup>244</sup>Cm in isotopic power sources for space and other applications calls for detailed nuclear data on their formation and fission properties. This type of information is also needed for detailed burn-up calculations and will inevitably contribute to a general knowledge and understanding of the actinides and their fission processes.

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# MODELE CONTINU POUR LE RALENTISSEMENT INELASTIQUE DES NEUTRONS RAPIDES

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# Abstract — Résumé

CONTINUOUS MODEL FOR THE INELASTIC SLOWING-DOWN OF FAST NEUTRONS. Following up the preliminary studies on the approximate operator for inelastic scattering, presented at the Karlsruhe symposium in June 1966 organized by the European Atomic Energy Society, the authors study its application to treatment of the inelastic slowing-down of <sup>239</sup>Pu, either alone or associated with the elastic slowingdown of oxygen. <sup>239</sup> Pu and oxygen are represented by their detailed microscopic cross-sections. For pure <sup>239</sup> Pu, the errors introduced by using the continuous model are assessed from some significant parameters (direct spectra, slowing-down density,  $k_{eff}$ , etc.) and compared with the errors caused by transition to the classic multigroup convention. For the Pu/O<sub>2</sub> mixture, the authors study the validity of a joint model for the inelastic scattering of <sup>239</sup> Pu and the elastic scattering of oxygen in the inelastic slowing-down range. In the same range, they check the conservation of the fine structure of the flux produced by oxygen scatter resonances.

MODELE CONTINU POUR LE RALENTISSEMENT INELASTIQUE DES NEUTRONS RAPIDES. A la suite des études préliminaires sur l'opérateur approché de diffusion inélastique, présentées à Karlsruhe lors du colloque de juin 1966 organisé par la Société européenne d'énergie atomique, les auteurs ont étudié son application au traitement du ralentissement inélastique du <sup>239</sup> Pu, soit seul, soit associé au ralentissement élastique de l'oxygène. Le <sup>239</sup>Pu e t'oxygène sont représentés par leurs sections efficaces microscopiques détaillées. Pour le <sup>239</sup>Pu pur, on a évalué sur certaines grandeurs significatives (spectres directs, densité de ralentissement, k<sub>eff</sub>, etc.) les erreurs introduites par l'utilisation du modèle continu et on les a comparées aux erreurs introduites par le passage au formalisme multigroupe classique. Pour le mélange Pu-O<sub>2</sub>, on a étudié la validité d'un modèle commun de diffusion inélastique du <sup>239</sup>Pu et de diffusion élastique de l'oxygène dans le domaine de ralentissement inélastique. Dans ce même domaine, on a vérifié la conservation de la structure fine du flux provoquée par les résonances de diffusion de l'oxygène.

#### INTRODUCTION

Les calculs des milieux à neutrons rapides sont en général tributaires de la formulation multigroupe, principalement à cause de la structure complexe de l'opérateur exprimant la diffusion inélastique. Cette méthode a l'inconvénient de dissimuler les faits physiques prépondérants sous une masse de données numériques.

C'est pourquoi nous avons proposé [1] un modèle simplifié permettant de traiter l'énergie comme une variable continue et susceptible donc de conduire à une meilleure compréhension des milieux multiplicateurs à neutrons rapides. Nous avons considéré des milieux homogènes où la dépendance vis-à-vis de l'espace est représentée par un mode fondamental (laplacien), mais le modèle proposé pourrait être étendu au traitement de problèmes plus complexes.

# 1. ETUDE DU MODELE CHOISI POUR LA DIFFUSION INELASTIQUE

Considérons le bilan neutronique dans un milieu infini avec une distribution homogène de sources:

$$\int_{E}^{\infty} P(E^{\dagger} \rightarrow E) \Sigma_{s}(E^{\dagger}) \Phi(E^{\dagger}) dE^{\dagger} - \Sigma_{t}(E) \Phi(E) + S(E) = 0$$

 $P(E' \rightarrow E)$  représente la probabilité élémentaire de transition. Le modèle est défini par un noyau synthétique de la forme:

$$\tilde{P}(E' \rightarrow E) = f(E')g(E) \quad \text{si } E' \ge E$$
  
= 0 si E' < E (1)

Pour obtenir ces fonctions nous avons imposé les deux conditions suivantes:

1) conservation de la section totale de diffusion inélastique:

$$\int_{0}^{E'} \widetilde{P}(E^{\dagger} \rightarrow E) dE = f(E^{\dagger}) \int_{0}^{E'} g(E) dE = 1$$
(2)

2) égalité de l'action des opérateurs exact et approché sur un certain spectre de référence  $\Phi_0(E)$ :

$$\int_{E}^{\infty} \widetilde{\mathbf{P}}(\mathbf{E}^{\dagger} \to \mathbf{E}) \Sigma_{s}(\mathbf{E}^{\dagger}) \Phi_{0}(\mathbf{E}^{\dagger}) d\mathbf{E}^{\dagger} = \int_{E}^{\infty} \mathbf{P}(\mathbf{E}^{\dagger} \to \mathbf{E}) \Sigma_{s}(\mathbf{E}^{\dagger}) \Phi_{0}(\mathbf{E}^{\dagger}) d\mathbf{E}^{\dagger} = \rho(\mathbf{E})$$
(3)

Par suite de la normalisation (2), on arrive à caractériser un corps par une seule fonction de l'énergie, soit h(E) = 1/f(E), telle que:

$$\widetilde{P}(E' \to E) = \frac{h'(E)}{h(E')}$$
(4)

h(E) est une fonction positive, partant de la valeur 0 pour E = 0 et croissant avec l'énergie.

L'expression analytique de h(E) obtenue par les équations (2) et (3) est:

avec

$$\omega(\mathbf{E}^{\dagger}) = \Sigma_{\mathbf{s}}(\mathbf{E}^{\dagger})\Phi_{0}(\mathbf{E}^{\dagger})$$
(5)

$$h(E) = C \cdot \exp \int_{E}^{\infty} \frac{\rho(E^{\prime}) dE^{\prime}}{\int_{E^{\prime}}^{\infty} [\omega(E^{\prime}) - \rho(E^{\prime})] dE^{\prime}}$$
(6)

h(E) est définie à une constante multiplicative près,  $h(\infty)$  est donc fixée arbitrairement.

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Pour obtenir une fonction continue de l'énergie, il est nécessaire de posséder des sections efficaces et des probabilités de transfert pour toute valeur de l'énergie. Actuellement, les connaissances disponibles sont des valeurs numériques prises par les quantités ci-dessus, en un nombre discret N d'énergies données. (Nous avons employé les sections efficaces microscopiques et les probabilités de transfert anglaises [2, 3].)

Nous devons donc transposer les équations continues pour obtenir un nouveau formalisme utilisable à partir d'un nombre fini de données numériques.

Les données consistent en:

1) une échelle d'énergies de départ, soit E<sub>i</sub> une de ces énergies,

2) la valeur numérique de la section efficace inélastique pour chacune de ces énergies, soit  $\sigma_i\equiv\sigma(E_i),$ 

 la valeur numérique (calculée à partir des données) de la probabilité de transfert d'une énergie E<sub>j</sub> de départ à une énergie E<sub>i</sub> d'arrivée, soit

$$P_{i,i} \equiv P(E_i \rightarrow E_i)$$

De plus, soit  $\varphi_j \equiv \Phi_0(E_j)$  la valeur numérique prise par le flux d'ajustement à l'énergie  $E_j$ .

Soit  $\mu_j$  le coefficient d'intégration<sup>1</sup> à l'énergie  $E_j$  apparaissant dans l'expression numérique des intégrales.

Nous nous sommes fixé une échelle d'énergies d'arrivée identique à l'échelle donnée d'énergies de départ et, dans ce nouveau formalisme,  $\rho(\mathbf{E})$  est donc calculé pour chaque énergie de l'échelle, soit  $\rho_i \equiv \rho(\mathbf{E}_i)$ .

La probabilité approchée qui, en continu, s'écrivait:

$$P(E' \rightarrow E) = f(E')g(E)$$

devient

$$\widetilde{\mathbf{P}}(\mathbf{E}_{i} \rightarrow \mathbf{E}_{i}) \equiv \widetilde{\mathbf{P}}_{i,i} = \mathbf{f}_{i} \mathbf{g}_{i}$$

 $\tilde{P}_{i,j} = f_{i,j}g_{i,j}$  s'obtient par la résolution du système suivant:

$$\sum_{i \leq j} \mu_i f_j g_i = 1$$
 (7)

$$\sum_{j=i}^{N} \mu_{j} f_{j} g_{i} \sigma_{j} \varphi_{j} = \sum_{j=i}^{N} \mu_{j} \bar{P}_{j,i} \sigma_{j} \varphi_{j} = \rho_{j}$$
(8)

Pour des commodités de calcul, on introduit la fonction  $h_i = 1/f_i$ ; la relation (7) donnant alors  $\mu_i g_i = h_i - h_{i-1}$ , les  $h_i$  sont obtenus par ré-

<sup>&</sup>lt;sup>1</sup> La conservation des neutrons doit être respectée; ces coefficients sont donc définis par la relation:  $\sum_{i \leq i} \mu_i P_{j,i} = 1.$ 

currence avec  $h_i = h_{i+1} - \rho_{i+1} \mu_{i+1} / \left( \sum_{j \ge i+1} \mu_j \omega_j / h_j \right)$ . En posant  $h_N = 1$  et

 $\omega_j = \sigma_j \varphi_j$ ,  $\tilde{P}_{j,i}$  s'écrit:

$$\widetilde{P}_{j,i} = \frac{h_i - h_{i-1}}{\mu_i h_i}$$

On retrouye bien l'équivalence dans le nouveau formalisme adopté .de l'expression théorique  $\tilde{P}(E' \rightarrow E) = h'(E)/h(E')$ .

Les éléments indiqués ci-dessus nous permettent d'effectuer à partir des données microscopiques, soit un calcul «exact», qui servira de référence, avec les  $P_{i,i}$ , soit un calcul approché avec les  $\tilde{P}_{i,i}$ .

# 2. COMPARAISON AVEC LE FORMALISME MULTIGROUPE CLASSIQUE

Les calculs actuels de neutronique se font à l'aide du formalisme multigroupe; les sections multigroupes s'obtiennent en effectuant des moyennes sur des sections microscopiques détaillées.

Nous avons fait un calcul exact avec des données microscopiques; nous allons maintenant créer à partir de ces données un formalisme multigroupe et comparer, par rapport aux résultats exacts obtenus avec les sections microscopiques, les erreurs introduites par l'utilisation du formalisme multigroupe à celles introduites par l'utilisation du modèle.

Pour que l'ensemble des études soit cohérent, il faut conserver pour la création des données multigroupes:

1) le flux d'ajustement utilisé pour la fabrication du modèle,

2) les coefficients d'intégration  $\mu_i$ .

Les données multigroupes dans le groupe [i] s'obtiennent alors par les équations suivantes:

 $\Phi_{0_{[i]}}$ : flux d'ajustement pour le groupe [i]

$$\Phi_{0[i]} = \int_{[i]} \Phi_{0}(\mathbf{E}') d\mathbf{E}' = \sum_{i \in [i]} \mu_{i} \varphi_{i}$$

 $\Sigma_{[i]}$ : section efficace pour le groupe [i]

$$\Sigma_{[i]} = \frac{\int \sigma(\mathbf{E'}) \Phi_0(\mathbf{E'}) d\mathbf{E'}}{\int \int \Phi_0(\mathbf{E'}) d\mathbf{E'}} = \frac{\sum_{i \in [i]} \mu_i \sigma_i \varphi_i}{\sum_{i \in [i]} \mu_i \varphi_i}$$

 $\Sigma_{[i] \rightarrow [i]}$ : section de transfert inélastique

$$\Sigma_{[i] \rightarrow [j]} = \frac{\int d\mathbf{E} \int \mathbf{P}(\mathbf{E}' \rightarrow \mathbf{E}) \sigma(\mathbf{E}') \Phi_0(\mathbf{E}') d\mathbf{E}'}{\int \int \Phi_0(\mathbf{E}') d\mathbf{E}'}$$
$$= \frac{\sum_{i \in [i]} \mu_i \sigma_i \varphi_i \sum_{i \in [j]} \mu_j \mathbf{P}_{i,j}}{\sum_{i \in [i]} \mu_i \varphi_i}$$

# 3. RESULTATS NUMERIQUES

Pour compléter les comparaisons, on peut, ainsi qu'il l'a été montré dans des études préliminaires [1], établir un modèle approché de diffusion inélastique dans le cadre du formalisme multigroupe classique. Soit  $\tilde{P}_{[j]\rightarrow[i]}$  la probabilité de transfert du groupe [j] au groupe [i], on peut la mettre sous la forme  $\tilde{P}_{[j]\rightarrow[i]} = F_j \sigma_i$ . Cette opération est indépendante des calculs faits à l'aide des sections microscopiques, elle permet de simplifier les calculs à l'intérieur du formalisme multigroupe.

Le schéma ci-dessous montre les différentes opérations effectuées:

Sections microscopiques  $\Rightarrow$  sections microscopiques approchées

$$(P(E_j \rightarrow E_i)) \qquad \qquad (\tilde{P}(E_j \rightarrow E_i)) \\ \downarrow \\ ons multigroupes \implies sections multigroupes appro}$$

Sections multigroupes  $\implies$  sections multigroupes approchées  $(\widetilde{P}_{[i] \rightarrow [i]})$   $(\widetilde{P}_{[i] \rightarrow [i]})$ 

Dans chaque formalisme (microscopique et multigroupe), nous avons calculé le flux, le facteur de multiplication effectif ( $k_{eff}$ ), la moyenne du temps de génération ( $\tau$ ), en utilisant successivement P et  $\tilde{P}$ . Pour différents laplaciens géométriques, nous avons résolu l'équation du bilan de neutrons dans l'approximation de la diffusion, écrite ici sous sa forme théorique continue:

$$\frac{1}{v(t)} \frac{d\Phi(E, t)}{dt} = -D(E) B^2 \Phi(E, t) + \int_{E}^{\infty} \Sigma_s(E' \to E) \Phi(E', t) dE' - \Sigma_t(E) \Phi(E, t) + S(E, t)$$
(9)



FIG.2. A justement sur fission: erreurs relatives sur  $k_{C}$  (Pu).

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Si l'on prend comme source  $S(E, t) = \chi(E) \delta(t)$ ,  $\chi(E)$  étant le flux de fission,  $k_{eff}$  et  $\tau$  sont obtenus par les intégrales [1]

$$\mathbf{k}_{eff} = \int_{0}^{\infty} \nu(\mathbf{E}) \Sigma_{f}(\mathbf{E}) d\mathbf{E} \int_{0}^{\infty} \Phi(\mathbf{E}, t) dt$$

$$\tau = \int_{0}^{\infty} \nu(\mathbf{E}) \Sigma_{f}(\mathbf{E}) d\mathbf{E} \int_{0}^{\infty} t \Phi(\mathbf{E}, t) dt$$
(10)

Ces équations doivent être transposées dans les deux formalismes et calculées chaque fois avec les probabilités de transfert exactes et approchées.

Les résultats numériques obtenus pour un bloc de plutonium pur sont exposés sous forme de figures.

3.1. Ajustement sur le flux de fission

La figure 1 donne la courbe h(E) interpolée par rapport aux points discrets  $h_i$ .

Dans les deux figures suivantes (fig. 2 et 3), on a calculé les valeurs de  $k_{eff}$  et de  $\tau$  pour différents laplaciens géométriques:

B <sup>2</sup> (m <sup>-2</sup> )	= 0,	3	0,25	0,2	0,15	0,1	0,05
k <sub>eff</sub>	= 0, 34	£306 0	, 40051	0,48119	0,60287	0,80785	1,22920
$10^8~ au$ (sec	) = 0,20	929 0	,23891	0,27868	0,33517	0,42255	<b>0,</b> 57929

Soient

 $k_c$  et  $\tau_c$  les valeurs de  $k_{eff}$  et de  $\tau$  calculées directement à partir des données microscopiques,

 $\tilde{k_c}$  et  $\tilde{\tau_c}$  les valeurs de  $k_{eff}$  et de au calculées par le modèle à partir des mêmes données,

 $k_m$  et  $au_m$  les valeurs de  $k_{eff}$  et de au en multigroupe,

 $\tilde{k}_m$  et  $\tilde{\tau}_m$  les valeurs de k<sub>eff</sub> et de  $\tau$  en multigroupe approché par le modèle.

On a (fig. 2):

$$\frac{\Delta k_{c}}{k_{c}} = \frac{\widetilde{k_{c}} - k_{c}}{k_{c}}$$

$$\frac{\Delta k_{mc}}{k_{c}} = \frac{k_{m} - k_{c}}{k_{c}}$$

$$\frac{\Delta k_{m}}{k_{c}} = \frac{\Delta k_{mc}}{k_{c}} + \frac{\widetilde{k}_{m} - k_{m}}{k_{c}}$$







FIG.5. A justement sur flux exact,  $B^2 = 0, 1 \text{ m}^{-2}$ : erreur relative sur k<sub>c</sub> (Pu).

et (fig. 3):

$$\frac{\Delta \tau_{\rm c}}{\tau_{\rm c}} = \frac{\tilde{\tau}_{\rm c} - \tau_{\rm c}}{\tau_{\rm c}}$$

$$\frac{\Delta \tau_{\rm mc}}{\tau_{\rm c}} = \frac{\tau_{\rm m} - \tau_{\rm c}}{\tau_{\rm c}}$$

$$\frac{\Delta \tau_{\rm m}}{\tau_{\rm c}} = \frac{\Delta \tau_{\rm mc}}{\tau_{\rm c}} + \frac{\tilde{\tau}_{\rm m} - \tau_{\rm m}}{\tau_{\rm c}}$$

La figure 4 donne les courbes  $\Phi(E)$ , flux exact caractéristique du milieu pour  $B^2 = 0, 1 \text{ m}^{-2}$ ,  $\tilde{\Phi}(E)$ , flux approché caractéristique du milieu pour  $B^2 = 0, 1 \text{ m}^{-2}$  et  $\chi(E)$ , flux de fission.

# 3.2. Ajustement sur flux caractéristique du milieu obtenu pour $B^2 = 0, 1 m^{-2}$

Ce flux est calculé directement avec (9) à partir des données microscopiques. Les calculs suivants sont faits par la méthode «continue» proposée (fig. 5 et 6):  $\Delta k_c/k_c = (\tilde{k}_c - k_c)/k_c; \Delta \tau_c/\tau_c = (\tilde{\tau}_c - \tau_c)/\tau_c$ .

On a calculé les flux exact et approché caractéristiques du milieu pour les différents laplaciens et représenté les erreurs relatives sur les flux exacts pour B<sup>2</sup> = 0,05, 0,15, 0,20 m<sup>-2</sup>. Pour B<sup>2</sup> = 0,1,  $\Delta\Phi(E)/\Phi(E)$  = 0 puisque l'ajustement est fait sur ce

Pour  $B^2 = 0, 1$ ,  $\Delta \Phi(E)/\Phi(E) = 0$  puisque l'ajustement est fait sur ce flux même. La figure 7 donne les courbes  $\Delta \Phi(E)/\Phi(E) = (\tilde{\Phi}(E) - \Phi(E))/\Phi(E)$ pour  $B^2 = 0,05$ , 0,15 et 0,20 m<sup>-2</sup>.









# 4. MELANGE Pu-O<sub>2</sub>

Nous savons que, pour des noyaux lourds, la diffusion élastique peut être représentée de manière satisfaisante par un modèle factorisé[4]. Nous allons voir, dans le domaine d'énergies supérieures à 10 keV où le ralentissement inélastique est associé au ralentissement élastique, si un









 $\begin{array}{l} ---- & \text{flux de fission} \\ ----- & \Phi(E) \\ ----- & \widetilde{\Phi}(E). \end{array}$ 



FIG. 10. A justement sur flux exact,  $B^2 = 0.05 \text{ m}^{-2}$ : erreur relative sur k<sub>c</sub> (Pu-O<sub>2</sub>).



FIG. 11. A justement sur flux exact: erreurs relatives sur les flux directs (Pu-O2).

<del></del>	$B^2 = 0,05 \text{ m}^{-2}$		
	$B^2 = 0,07 \text{ m}^{-2}$	B <sup>2</sup> = 0, 10 m <sup>-2</sup>	

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modèle unique permet une bonne représentation de l'ensemble des phénomènes de diffusion. Le ralentissement élastique engendré par le plutonium est négligeable par rapport à son ralentissement inélastique, nous étudions donc un modèle représentant la diffusion inélastique du plutonium et la diffusion élastique de l'oxygène.

Nous adoptons:

1) une échelle d'énergies commune aux deux corps; les résultats numériques portant sur des bilans neutroniques, nous avons pu lisser les courbes représentant les résonances de l'oxygène et conserver l'échelle moins détaillée du plutonium;

2) le formalisme employé pour le plutonium seul; nous utilisons des notations équivalentes à celles qui ont été définies précédemment, les indices supplémentaires e correspondant à la diffusion élastique (par ex.  $\sigma_{e_i}$ ) et i correspondant à la diffusion inélastique (par ex.  $\sigma_{i_i}$ ).

 $\tilde{P}_{i,i} = f_i g_i$  est solution du système

$$\sum_{i \le j} \mu_i f_j g_i = 1$$

$$\sum_{j=i}^{N} \mu_{j} f_{j} g_{i} [\sigma_{e_{j}} + \sigma_{i_{j}}] \varphi_{j} = \sum_{j=i}^{N} \mu_{j} [\sigma_{e_{j}} P_{e_{j,i}} + \sigma_{i_{j}} P_{i_{j,i}}] \varphi_{j} = \rho_{j}$$

En résolvant l'équation du bilan neutronique écrite pour le mélange Pu-O<sub>2</sub> et transposée dans le nouveau formalisme, nous avons calculé le flux et le facteur de multiplication effectif ( $k_{eff}$ ).

Le ralentissement est, dans l'équation, successivement représenté par:

1) les matrices exactes:

$$\sum_{j=i}^{N} \mu_{j} \left[ \sigma_{e_{j}} P_{e_{j,i}} + \sigma_{i_{j}} P_{i_{j,i}} \right] \varphi_{j}$$

2) la matrice approchée:

$$\sum_{j=i}^{N} \mu_{j} f_{j} g_{i} [\sigma_{e_{j}} + \sigma_{i_{j}}] \varphi_{j}$$

Les calculs sont effectués avec les données microscopiques; nous n'avons pas fait le calcul des sections multigroupes correspondantes.

Les  $k_{eff}$  et les flux ont été calculés pour différentes valeurs du laplacien:

B <sup>2</sup> (m <sup>-2</sup> )	0,05	0,07	0,08	0,10	0,12	0,15
k <sub>eff</sub>	0,91772	0,72684	0,65840	0,55411	0,47843	0,39719

(11)





# 4.1. A justement sur flux de fission pour $E \ge 1$ MeV et constante pour $E \le 1$ MeV

La figure 8 donne  $\Delta k_c/k_c = (\tilde{k}_c - k_c)/k_c$  avec les mêmes notations que précédemment. La figure 9 donne  $\Phi(E)$  (flux exact caractéristique du milieu pour B<sup>2</sup> = 0,05 m<sup>-2</sup>),  $\tilde{\Phi}(E)$  (flux approché caractéristique du milieu pour B<sup>2</sup> = 0,05 m<sup>-2</sup>) et  $\chi(E)$  (flux d'ajustement).

# 4.2. Ajustement sur flux caractéristique du milieu pour $B^2 = 0,05 \text{ m}^{-2}$

La figure 10 donne  $\Delta k_c/k_c = (\tilde{k}_c - k_c)/k_c$  et la figure 11 la représentation des erreurs relatives obtenues sur le flux exact pour différents laplaciens:  $\Delta \Phi(E)/\Phi(E) = (\tilde{\Phi}(E) - \Phi(E))/\Phi(E)$  pour B<sup>2</sup> = 0,07, 0,08 et 0,1 m<sup>-2</sup>, et  $\Delta \Phi(E)/\Phi(E) = 0$  pour B<sup>2</sup> = 0,05 m<sup>-2</sup>.

# 5. DIFFUSION ELASTIQUE

Dans un mélange  $Pu-O_2$ , pour des énergies élevées apparaît une structure fine du flux provoquée par les résonances de diffusion de l'oxygène.

Prenant simplement de l'oxygène en présence d'un corps d'absorption constante, nous avons calculé le flux caractéristique de ce milieu en employant successivement les probabilités de transfert exacte et approchée de l'oxygène. ( $\tilde{P}_{j,i} = (h_i - h_{i-1})/\mu_i h_j$ ,  $h_i$  calculé par la méthode indiquée pour le plutonium en prenant comme flux de référence le flux de fission.) Nous avons pu vérifier une conservation satisfaisante de cette structure fine. Le flux exact ( $\Phi(E)$ ) et le flux approché ( $\tilde{\Phi}(E)$ ) en milieu infini sont reproduits à la figure 12.

# CONCLUSIONS - ACTIVITES FUTURES

Les résultats obtenus montrent que le modèle étudié peut constituer un outil commode pour l'étude de nombreux problèmes où une connaissance détaillée du spectre de neutrons est nécessaire.

Parmi les applications intéressantes que nous envisageons maintenant nous citerons:

- Etude de bilan de neutrons en milieu infini, comparaison de milieux
   <sup>235</sup>U et Pu, comparaison avec des calculs classiques (multigroupes et Elmoe) sur des compositions représentatives des milieux «Phénix»:
- Etude de la dépendance spatiale de la structure fine énergétique du spectre: milieux hétérogènes, problèmes d'interface;
- Etude de problèmes multizones, soit directement, soit par utilisation conjointe du modèle continu et du formalisme multigroupe classique.

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# DISCUSSION

T.D. BEYNON: This is a very interesting technique. Looking at the physics of it, one is reminded of the Weisskopf evaporation formula for inelastic scattering, where the inelastic kernel is regarded as degenerate. It is well known that the evaporation model gives quite good results for heavy-mass isotopes, where there is a dense target structure, but for light isotopes the kernel is, of course, not so good. Have you tried out your method on slowing down in a medium moderator like iron?

M. PUJOL: No, we have only done calculations for plutonium and plutonium oxide.

T.D. BEYNON: Referring to the evaporation model once more, I suspect that, in fact, you are finding effective evaporation temperatures by using what you call a reference spectrum. I would hazard a guess that the method described in your paper would not be so effective for an isotope of medium mass.
# IMPRECISIONS DES

PARAMETRES CARACTERISTIQUES D'UN REACTEUR RAPIDE DE PUISSANCE DUES AUX INCERTITUDES ACTUELLES CONCERNANT LES DONNEES NEUTRONIQUES Leur évaluation, et leur diminution à partir des expériences critiques

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#### Abstract — Résumé

INACCURACIES IN THE CHARACTERISTIC PARAMETERS OF A FAST POWER REACTOR DUE TO THE PREVAILING UNCERTAINTIES IN THE BASIC NEUTRON DATA: THEIR EVALUATION AND THE EXTENT TO WHICH THEY CAN BE REDUCED BY CRITICAL EXPERIMENTS. The authors first determined the inaccuracies in the characteristic parameters of a fast reactor due to uncertainties in the basic neutron data, using a generalized perturbation method. The magnitude of these uncertainties then led them, as a second step, to use the results of critical experiments to improve the accuracy of the calculated forecasts and thereby possibly improve the cross-sections themselves.

The first method, described by Baker, simply provides an empirical formula for correcting the calculated critical mass and has been directly applied to the Cadarache data. By the second method (the BARRAKA code) one obtains a set of corrections to the cross-sections considered to be of major importance after making adjustments as between the experimental values and those calculated from the Cadarache data for several parameters measured in a large number of critical fast assemblies.

IMPRECISIONS DES PARAMETRES CARACTERISTIQUES D'UN REACTEUR RAPIDE DE PUISSANCE DUES AUX INCERTITUDES ACTUELLES CONCERNANT LES DONNEES NEUTRONIQUES: LEUR EVALUATION, ET LEŪR DIMINUTION A PARTIR DES EXPERIENCES CRITIQUES. Dans une première étape, les auteurs ont déterminé les imprécisions des paramètres caractéristiques d'un réacteur rapide dues aux incertitudes concernant les données neutroniques de base à l'aide d'une méthode de perturbation généralisée. L'importance de ces imprécisions les a conduits, dans une seconde étape, à utiliser les résultats des expériences critiques pour améliorer la précision des prévisions par calcul et éventuellement les sections efficaces elles-mêmes,

La première méthode (celle de Baker) fournit simplement une formule empirique pour corriger la masse critique calculée et a été directement appliquée au formulaire Cadarache. La seconde méthode (code BARRAKA) donne au contraire un ensemble de corrections pour les sections efficaces jugées prépondérantes après ajustement entre les valeurs expérimentales et celles calculées par le formulaire Cadarache de plusieurs paramètres mesurés dans un grand nombre d'assemblages rapides critiques.

#### INTRODUCTION

Le premier but de cette étude est de déterminer l'influence des incertitudes actuelles concernant les données neutroniques de base sur les paramètres caractéristiques (masse critique, taux de surgénération, temps de vie ...) d'un grand réacteur rapide de puissance.

En utilisant la méthode de perturbation généralisée décrite par Usachev [1-3], nous avons pu localiser parmi l'ensemble des sections efficaces les sources d'erreurs les plus importantes et estimer une erreur globale maximale par élément pour chaque paramètre (par exemple  $\pm 4\%$  pour la réactivité due au <sup>239</sup>Pu). En plus de leur intérêt propre, ces résultats nous permettent de formuler pour les expérimentateurs un ensemble cohérent de demandes de mesure, avec les précisions souhaitées et les degrés d'urgence.

Les imprécisions importantes constatées sur les paramètres caractéristiques nous ont conduits à essayer de tenir compte des résultats des expériences critiques pour améliorer la précision des prévisions par calcul et éventuellement améliorer les sections efficaces elles-mêmes. Dans ce but, deux méthodes sont envisagées:

La méthode décrite par Baker [4] fournit une formule empirique fonction de la composition permettant de corriger la masse critique calculée par un jeu de constantes donné. Les coefficients de régression qui interviennent dans cette formule sont caractéristiques du jeu et déterminés par ajustement par moindres carrés entre les valeurs calculées par le jeu et mesurées du coefficient de multiplication d'un grand nombre d'expériences critiques. Cette méthode a simplement été programmée dans le code BARBAK et appliquée au «jeu Cadarache» [5] sur 34 assemblages. Nous estimons avoir réduit l'écart-type sur la réactivité de 1,6% à 0,3%.

Une seconde méthode (code BARRAKA) consiste à ajuster les sections efficaces de façon à obtenir le meilleur accord entre tous les paramètres calculés par un jeu donné et mesurés dans les expériences critiques. Des recherches dans cette direction ont déjà été entreprises par Pendlebury et al. [6], Pazy et al. [7], et également par Gandini et al. [8]. Toutes relient la variation d'un paramètre d'un assemblage à la variation d'une section efficace par une méthode de perturbation généralisée.

Dans ce mémoire, les sections efficaces choisies sont celles dont l'influence a été jugée prépondérante d'après l'étude précédente; elles ne peuvent varier que dans les limites des incertitudes actuelles estimées dans cette étude. Nous avons imposé que le nombre de paramètres mesurés soit nettement supérieur au nombre de sections efficaces à ajuster. L'imprécision sur un paramètre mesuré est simplement prise en compte par l'intermédiaire d'un poids. Le fait d'utiliser un système surabondant permet de s'abstenir de toute condition supplémentaire arbitraire sur les variations des sections efficaces à l'intérieur des contraintes choisies.

Les tendances qui se dégagent des résultats préliminaires (tendances indépendantes des poids attribués aux différents paramètres) conduiraient, pour le jeu Cadarache, à augmenter la capture du <sup>239</sup>Pu et du <sup>235</sup>U et à diminuer la fission et le  $\nu$  de ces deux éléments et la capture du <sup>238</sup>U.

Il n'est pas question d'utiliser cette méthode comme moyen d'évaluation automatique, ni de modifier actuellement les données microscopiques de base. Toutefois, la méthode nous permet dans l'immédiat d'améliorer la précision de beaucoup de paramètres calculés et pourra peut-être, une fois au point, faciliter la tâche des évaluateurs.

# 1. IMPRECISIONS DES PARAMETRES CARACTERISTIQUES D'UN REACTEUR RAPIDE DE PUISSANCE DUES AUX INCERTITUDES ACTUELLES CONCERNANT LES DONNEES NEUTRONIQUES

Les incertitudes sur les paramètres caractéristiques d'un réacteur rapide proviennent de deux sources: les erreurs dues aux méthodes de calcul, et les erreurs dues aux imprécisions dans les données neutroniques multigroupes. Notre but est uniquement de déterminer les incertitudes dues aux erreurs du second type, dont l'amplitude est à notre avis prépondérante.

# 1.1. Incertitudes actuelles dans les données neutroniques

Deux types essentiels d'incertitudes peuvent intervenir sur les sections efficaces effectives des éléments entrant dans la composition d'un réacteur:

- Les erreurs dues aux données microscopiques provenant, soit des erreurs mentionnées par un expérimentateur (erreur statistique), soit des erreurs systématiques entre les valeurs mesurées par différents expérimentateurs, soit des imprécisions des sections efficaces calculées par différents modèles théoriques dans les domaines où les mesures n'existent pas;

- Les erreurs dues aux méthodes de passage des données microscopiques aux sections efficaces multigroupes.

Seules les premières erreurs ont été évaluées, en distinguant trois catégories d'éléments:

Eléments lourds : <sup>238</sup>U, <sup>239</sup>Pu, <sup>240</sup>Pu

Eléments intermédiaires: Fe, Ni, Na

Elément léger : O.

Etant donné la faible importance de la diffusion élastique par les éléments lourds dans le ralentissement, cette section efficace n'a pas été étudiée. De même, la diffusion inélastique par l'oxygène n'est pas considérée.

Les incertitudes maximales sur toutes les sections efficaces et le paramètre  $\nu$  ont été établies dans le découpage en énergie du jeu russe[9]. S'il existe une part d'arbitraire dans le choix de ces incertitudes maximales, elles donnent cependant une bonne représentation des erreurs dont sont entachées actuellement les données neutroniques de base. Pour les sept éléments cités, le détail de ces incertitudes est donné dans les rapports de Barré [10, 11].

# 1.2. Relation entre incertitudes sur les données de base d'une part et sur les paramètres d'un réacteur d'autre part

# 1.2.1. Méthode

Dans des études similaires, Moorhead [12] et Greebler et al.[13] ont utilisé des calculs directs. Nous employons une méthode de perturbation généralisée décrite par Usachev [1-3].

En effet, l'influence des incertitudes sur les sections efficaces étant faible, une méthode directe qui donne la variation d'un paramètre à partir de la différence de deux résultats nécessite une grande précision de calcul. Une méthode de perturbation apporte pour la même précision un gain de temps et une meilleure compréhension des phénomènes physiques en définissant une expression analytique de cette variation. La méthode d'Usachev donne la variation de tout paramètre caractéristique qui s'exprime sous forme d'un rapport de deux fonctionnelles bilinéaires du flux et du flux adjoint, variation due à toute perturbation d'une donnée neutronique dans le réacteur, par exemple une section efficace. Le réacteur est ramené à l'état critique après la perturbation par variation de l'enrichissement. Cette méthode a été appliquée dans l'approximation de la diffusion et à zéro dimension dans le code PERTUS.

# 1.2.2. Résultats

Les six paramètres caractéristiques étudiés sont le coefficient de multiplication (k), le taux de surgénération interne (TRI), le temps de vie (l), le spectre caractérisé par le rapport du flux au-dessus de 1,4 MeV sur le flux total (R4), le coefficient sodium décomposé en effet dégradation (CNAD) et effet capture (CNAC).



FIG.1. Imprécisions sur k dues aux incertitudes des sections efficaces, dk/k = f(E), en pcm.

Comme modèle d'un grand réacteur rapide de puissance, nous avons choisi un réacteur à combustible mixte  $PuO_2 \cdot UO_2$ ;<sup>1</sup> le volume critique du cœur est de 1200 litres. A titre d'exemple, la variation en fonction de l'énergie de l'imprécision sur le coefficient de multiplication due aux incertitudes concernant les sections efficaces dont l'influence est prépondérante est représentée dans la figure 1. Ces imprécisions sur les paramètres caractéristiques, calculées initialement à 24 groupes, sont condensées pour la clarté des résultats en quatre bandes d'énergie. Le tableau I donne par exemple cette présentation condensée pour le paramètre k.

<sup>&</sup>lt;sup>1</sup> Compositions atomiques en  $10^{24}$  at. cm<sup>-3</sup>: <sup>239</sup>Pu:0,112 · 10<sup>-2</sup>; <sup>240</sup>Pu:0,295 · 10<sup>-3</sup>; <sup>238</sup>U:0,672 · 10<sup>-2</sup>; acier:0,225 · 10<sup>-1</sup>; O:0,164 · 10<sup>-1</sup>; Na:0,617 · 10<sup>-2</sup>.

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Elément	Energie	Fission	Capture	Transport	ν	Diffusion élastique	Diffusion inélastique
	1,4 - 10,5 MeV 0.0465 - 1.4 MeV	+ 719	- 34 -2120	+ 209 + 526	+ 351	-	- 694 - 146
238 U	0.215 - 46.5 keV	-	-2186	+ 46		۰.	-
	Therm 215 eV	-	- 36	+ 0	-	-	-
	1,4 - 10,5 MeV	+ 408	- 31	+ 33	+ 210	-	- 142
	0,0465 - 1,4 MeV	+ 2612	- 654	+ 110	+ 827		- 68
<sup>239</sup> Pu	0,215 - 46,5 keV	+ 2933	- 1447	+ 26	+ 292	-	-
	Therm 215 eV	+ 108	- 104	+ 0	+ 13	-	-
	1,4 - 10,5 MeV	+ 104	- 6	+ 13	+ 85	-	- 48
	0,0465 - 1,4 MeV	+ 208	- 313	+ 33	+ 132	-	- 16
240 Pu	0,215 - 46,5 keV	+ 5	- 417	+ 6	+ 3	-	-
	Therm 215 eV	-	- 18	+ 0	-	-	
	1,4 - 10,5 MeV	-	- 17	+ 350	-	- 23	- 587
	0,0465 - 1,4 MeV	-	- 226	+ 449	-	- 45	- 67
Fe	0,215 - 46,5 keV	-	- 355	+ 79	-	+ 20	-
	Therm 215 eV	-	- 3	+ 0	-	+ 0	-
	1,4 - 10,5 MeV	-	- 35	+ 28	-	- 5	- 128
	0,0465 - 1,4 MeV	-	- 94	+ 90	-	- 11	-
Ni	0,215 - 46,5 keV	-	- 47	+ 38	-	+ 3	-
	Therm, - 215 eV	-	- 0	+ 0	- '	+ 0	-
	1.4 - 10.5 MeV	-	- 3	+ 162	-	- 23	- 35
	0,0465 - 1,4 MeV	-	- 4	+ 151	-	- 41	- 13
Na	0,215 - 46,5 keV	-	- 5 ·	+ 18	-	+ 14	
	Therm, - 215 eV	-	- 0	+ 0	-	0	-
	1.4 - 10.5 MeV	-	- 129	+ 174	-	- 52	-
	0.0465 - 1.4 MeV	-	-	+ 739	-	- 265	-
0	0.215 - 46.5 keV	-	-	+ 89	-	- 12	-
	Therm - 215 eV	-	-	0		0	-
	Incin 210 ev.		l	ļ	l	ľ	ļ

# TABLEAU I. VARIATIONS DE LA REACTIVITE DUES AUX INCERTITUDES SUR LES SECTIONS EFFICACES: dk/k = f(+ $\Delta\sigma/\sigma)$ en pcm

Les principales conclusions se résument en quatre points:

a) Les données neutroniques dont les incertitudes sont jugées prépondérantes pour tous les paramètres sur la majeure partie du spectre d'énergie sont la capture et la fission du <sup>239</sup>Pu, et la capture du <sup>238</sup>U.

b) Si ces conclusions étaient prévisibles qualitativement, nous avons par contre chiffré l'importance relative des incertitudes de toutes ces sections sur chaque paramètre suivant l'énergie. Nous avons <u>estimé</u> l'imprécision globale sur chaque paramètre due aux erreurs sur toutes les sections dans tout le domaine d'énergie pour les trois éléments prépondérants (tableau II). Ces valeurs dépassent largement les marges souhaitées pour un projet.

c) L'influence des incertitudes sur les sections efficaces de tous les éléments, entre l'énergie thermique et 200 eV, puis entre 1,5 et 10 MeV, sur les paramètres étudiés est faible pour ce type de réacteur, à quelques exceptions près (diffusion inélastique dans le <sup>238</sup>U par exemple).

d) Pour ces paramètres, les précisions souhaitables pour un projet étant supposées de

± 0,6% sur k (soit 1% sur la masse critique par variation de l'enrichissement),

± 1% sur le taux de surgénération interne,

± 5% sur le temps de vie et le coefficient sodium,

pour les sections efficaces dont l'influence dans un domaine d'énergie est supérieure à l'une de ces limites, nous avons fait des demandes de mesure de priorité I dans la «Request List» de 1967 de l'EANDC. Les précisions demandées sont obtenues par simple proportionnalité.

Il serait souhaitable que ces demandes de mesures puissent être satisfaites, car les incertitudes existant sur des paramètres calculés aussi importants pour un projet de réacteur rapide que la masse critique ou le taux de surgénération doivent pouvoir être considérablement réduites.

# 2. UTILISATION DES RESULTATS DES EXPERIENCES CRITIQUES POUR DIMINUER LES IMPRECISIONS SUR LES PARAMETRES CALCULES DE REACTEUR

Il apparaît, d'après les résultats précédents, que la seule utilisation des données microscopiques mesurées ne nous permet pas de réaliser des calculs de réacteurs rapides avec une précision suffisante pour un projet. Il nous semble alors utile de tenir compte des paramètres mesurés dans les expériences critiques. Les buts des deux méthodes que nous employons dans ce sens sont opposés: la première méthode (décrite par

TABLEAU II. INCERTITUDES GLOBALES ESTIMEES DES PARAMETRES CARACTERISTIQUES POUR LES TROIS ELEMENTS PREPONDERANTS (%)

Elément	d k	<u>d TRI</u>	<u>d R4</u>	<u>d l</u>	d CNAD
	k	TRI	R4	l	CNAD
<sup>238</sup> U	± 4,6	± 12	± 10	± 17	± 50
<sup>239</sup> Рu	± 4	± 13	± 2	± 8	± 40
Fer	± 2	± 3	± 7	± 4	± 15

Baker [4]) fournit simplement une formule empirique qui permet de corriger la masse critique calculée par un jeu donné; la seconde méthode donne au contraire un ensemble de corrections pour les sections efficaces jugées prépondérantes, après ajustement sur tous les paramètres mesurés dans les assemblages.

#### 2.1. Méthode de Baker

#### 2.1.1. Principe

La base de cette méthode, directement appliquée à Cadarache dans le programme BARBAK, est l'ensemble des coefficients de multiplication k cobtenus par un jeu donné des modèles calculables d'un grand nombre M d'assemblages critiques rapides.

On suppose que l'écart entre la valeur calculée  $k^c$  et le coefficient de multiplication expérimental  $k^{ex} = 1$  peut se développer sous forme linéaire des compositions atomiques de N éléments présents dans le cœur des assemblages ( $^{239}$ Pu,  $^{235}$ U,  $^{238}$ U, etc.)

$$k^{c} - 1 = \sum_{i=1}^{N} a_{i} c_{i}$$

Les compositions atomiques sont normalisées comme suit:

c<sub>1</sub>, concentration en  $^{239}$ Pu, = 0,6 c<sub>2</sub>, concentration en  $^{235}$ U, = 1,0.

Les N coefficients de régression a<sub>i</sub>, caractéristiques du jeu, sont ceux qui minimisent la somme des carrés des résidus E<sup>2</sup> suivant la méthode habituelle des moindres carrés:

$$E^{2} = \sum_{j=1}^{M} \left( k^{c}_{j} - 1 - \sum_{i=1}^{N} a_{i} c_{ij} \right)^{2}$$

La méthode implique que le nombre M d'assemblages soit supérieur au nombre N d'inconnues  $a_i$ . Le fait que la régression obtenue soit significative est vérifié par un test en F de Student-Fisher, le seuil étant choisi à 1%. Le gain apporté par cette méthode peut être chiffré en comparant l'écart-type initial  $\sigma_I$  du coefficient de multiplication autour de sa valeur moyenne  $\bar{k}$  et l'écart-type après ajustement  $\sigma_F$ :

$$\sigma_{\rm I}^2 = \frac{\sum_{j=1}^{\rm M} (k^c_j - \overline{k})^2}{M - 1}$$
$$\sigma_{\rm F}^2 = \frac{E^2}{M - N - 1}$$

Elément	<sup>239</sup> Pu	<sup>235</sup> U	<sup>238</sup> U	Acier	Aluminium	Sodium	Oxygène	Carbone	Hydrogène	Cuivre
Coefficient de régression $a_i \times 10^4$	+ 406 ± 70	+ 183 . ± 19	- 37 ± 2	+ 15 ± 2	- 3 ± 3	+ 8 ±12	+ 14 ± 6	- 2 ± 2	- 77 ± 25	- 241 ± 63
Nombre d'assem- blages où l'é- lément est présent	4	30	33	34	14	5	3	13	4	3
Ecart-type	σI = 1,6	70				σ <sub>F</sub> = 0,3	70			
Analyse des	Rapport des variances : F = 81, 7Degrés de liberté $v_1 = 10$									
variances	Seuil à 1	%	: F = 3, 17 $\nu_2 = 23$							

TABLEAU III.COEFFICIENTS DE REGRESSION ET ECARTS-TYPES POUR LE JEUCADARACHE - METHODE BARBAK

#### 2.1.2. Application au jeu Cadarache

Base expérimentale. Les 34 assemblages calculés par ce jeu, choisis parmi les ZPR III, ZEBRA et VERA, comprennent 30 assemblages au  $^{235}$ U et quatre au  $^{239}$ Pu. Le calcul d'un assemblage se fait dans l'approximation de la diffusion sur un modèle homogène sphérique avec réflecteur. Le passage de l'assemblage critique expérimental au modèle critique simplifié pour le calcul nécessite plusieurs corrections de la masse critique expérimentale: correction expérimentale (barres, contour...), hétérogénéité, facteur de forme. Nous tenons compte des différences entre l'approximation de la diffusion et celle du transport (S4) en utilisant l'écart moyen donné par Baker [4] pour chaque assemblage. L'écart entre la masse critique calculée par le jeu et la masse critique expérimentale du modèle simplifié est alors traduit en variation de k par l'intermédiaire du facteur (dM/M)/(dk/k).

<u>Résultats</u>. Les coefficients de régression obtenus (tableau III) démontrent que les éléments <sup>239</sup>Pu et <sup>235</sup>U sont surréactifs et que le <sup>238</sup>U est sous-réactif comme dans la plupart des jeux [4].

Un ajustement important est réalisé pour l'ensemble des compositions (fig. 2): l'écart-type sur k passe de 1,6% à 0,3% après ajustement (pour le jeu FD2, l'écart-type final est de 0,45% [4]).

#### 2.1.3. Validité des résultats

Nous souhaitons utiliser cette méthode pour corriger la masse critique d'un grand réacteur rapide au  $^{239}$ Pu comprenant 20% de  $^{240}$ Pu. Or, seuls quatre assemblages au  $^{239}$ Pu sont utilisés; de plus, le combustible ne comprend en moyenne que 6% de  $^{240}$ Pu. Un ajustement effectué avec le  $^{240}$ Pu a donné un coefficient de régression positif, mais l'écart-type extrêmement important sur ce résultat lui ôte tout sens. D'après l'étude précédente (tableau I), pour un réacteur de ce type l'incertitude maximale sur k due aux 20% de  $^{240}$ Pu pourrait atteindre  $\pm$  1,5%.

Pour obtenir la valeur du paramètre k calculé qui se compare à la valeur expérimentale, il est nécessaire d'effectuer plusieurs corrections qui sont elles-mêmes entachées d'erreurs:

Hétérogénéité	:	±	0,2% sur k [14]
Facteur de forme	:	±	0,5% sur k [14]
Correction expérimentale	:	±	0,1% sur k [14]
Passage diffusion - S4	:	±	0,2% sur k

Pour le modèle homogène sphérique d'un même assemblage critique, il existe des désaccords qui peuvent atteindre  $\pm 1000$  pcm entre les valeurs globales de ces corrections évaluées par différents auteurs, Davey [14] et Meneghetti et al.[15] par exemple.

Les résultats obtenus avec les normalisations 0,5, 0,6 et 0,7 pour la composition du  $^{239}$ Pu (tout en conservant c<sub>2</sub> = 1,0) sont très voisins: ceci confirme également la conclusion de Baker.

Cette méthode, extrêmement attirante par sa simplicité, a été utilisée pour essayer de corriger la masse critique de l'assemblage MASURCA 1-A au <sup>239</sup>Pu [16] calculée par le jeu Cadarache:

Masse critiqu	e expérimentale <sup>2</sup>	170±0,6 kg
Masse critiqu	e calculée par le jeu <sup>3</sup>	141±5 kg
Masse critiqu	e corrigée par BARBAK	156±7 kg

Malgré le gain apporté par l'ajustement, le désaccord avec l'expérience demeure important. Une valeur plus faible de l'hétérogénéité en diminuerait l'amplitude.

Cet écart pourrait être attribué au petit nombre de réacteurs au <sup>239</sup>Pu utilisés dans l'ajustement. En effet, parmi les 34 réacteurs analysés initialement, supprimons un des 30 réacteurs à <sup>235</sup>U le plus mal calculé par le jeu (ZPR III-32).

La masse critique de cet assemblage corrigé par ce nouvel ajustement est en accord avec l'expérience:

Masse critique expérimentale 213 kg Masse critique calculée par le jeu 180 kg Masse critique corrigée BARBAK 217±5 kg

Il serait important de pouvoir disposer d'un plus grand nombre d'expériences critiques au plutonium pour corriger avec plus de garantie la masse critique calculée d'un réacteur rapide utilisant ce combustible.

# 2.2. Méthode BARRAKA

#### 2.2.1. Principe

A l'opposé de la méthode précédente, qui corrige la masse critique après le calcul, cette seconde méthode recherche, par ajustement entre tous les paramètres calculés par un jeu donné et mesurés dans les expériences critiques, l'ensemble des variations des sections efficaces qui donne le meilleur accord entre le calcul et l'expérience.

D'une part, nous disposons de A assemblages rapides critiques. Dans chaque assemblage a, P paramètres  $X^m$  (a, p) sont mesurés. Soient  $X^c$  (a, p) les valeurs de ces paramètres calculées par un jeu donné.

D'autre part, nous souhaitons ajuster des sections efficaces correspondant à R réactions, E éléments dans un découpage en énergie à G groupes. Cette méthode est caractérisée par les deux impératifs suivants: les variations des sections efficaces données par l'ajustement doivent être comprises entre les limites des incertitudes maximales actuelles estimées dans l'étude précédente (section 1); le nombre de paramètres mesurés doit être nettement supérieur au nombre de sections efficaces ajustées.

La variation relative dX/X d'un paramètre p mesuré dans un assemblage a est reliée à la variation relative  $d\sigma/\sigma$  d'une section efficace d'une

<sup>&</sup>lt;sup>2</sup> Masse critique équivalente en plutonium comprenant 8,5% de <sup>240</sup>Pu.

<sup>&</sup>lt;sup>3</sup> L'effet d'hétérogénéité est évalué actuellement à 2,5±0,5% sur k (KHAIRALLAH, A., ces comptes rendus, SM-101/57).

réaction r, un élément e et un groupe g par la méthode de perturbation généralisée au premier ordre déjà utilisée (1.2):

$$\frac{\mathrm{d}X^{\mathrm{c}}}{\mathrm{X}^{\mathrm{c}}}(\mathrm{a},\mathrm{p}) = \alpha(\mathrm{a},\mathrm{p},\mathrm{e},\mathrm{r},\mathrm{g}) \frac{\mathrm{d}\sigma}{\sigma}(\mathrm{e},\mathrm{r},\mathrm{g}) \tag{1}$$

Cette méthode de perturbation est également employée par d'autres auteurs (Gandini et al.[8]). Nous retenons les variations relatives des sections efficaces qui minimisent la somme des carrés des résidus  $E^2$ :

$$E^{2} = \sum_{a,p}^{A,p} W(a,p) \left[ \frac{X^{m}(a,p) - X^{aj}(a,p)}{X^{m}(a,p)} \right]^{2}$$
(2)

avec

$$X^{aj}(a,p) = X^{c}(a,p) \left[ 1 + \sum_{e,r,g}^{E,R,G} \alpha(a,p,e,r,g) \frac{d\sigma}{\sigma}(e,r,g) \right]$$

W(a, p) représente le poids attribué à la mesure d'un paramètre dans un assemblage.

Le problème se ramène à une minimisation avec contraintes. La résolution se fait par une méthode très simple de cheminement avec pas de variation variable pour chaque inconnue et éventuellement division de pas. En aucun cas la variation d'une section efficace ne peut dépasser en valeur absolue la borne évaluée, mais si elle l'atteint en cours d'ajustement, elle ne demeure pas nécessairement fixée à cette valeur.

Le fait d'imposer un système surabondant permet de s'abstenir de toute condition supplémentaire arbitraire sur les variations des sections efficaces à l'intérieur des contraintes choisies. Choisir parmi plusieurs solutions celle qui donne la somme minimale de variations pour l'ensemble des sections efficaces supprimerait la possibilité de tenir compte d'erreurs systématiques qui, à notre avis, peuvent encore exister ( $\alpha$  du <sup>239</sup>Pu par exemple). Le formalisme que nous utilisons ne donne aucune préférence aux valeurs de départ.

Le poids attribué à un paramètre mesuré se décompose en deux facteurs:

$$W(a, p) = \frac{K(p)}{x^2(a, p)}$$
 (3)

x(a, p) représente l'erreur sur un paramètre mesuré dans un assemblage, soit donnée par l'expérimentateur, soit évaluée. K(p) permet de faire évoluer arbitrairement l'importance relative des différents paramètres dans l'ajustement. Un des points délicats de la méthode est certainement le choix de ces poids. Cette méthode n'est valable que dans la mesure où les variations obtenues des sections efficaces sont indépendantes des poids relatifs choisis dans une gamme suffisamment étendue.

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2.2.2. Application au jeu Cadarache

Paramètres mesurés. Les cinq paramètres retenus dans cette première étude sont:

-le coefficient de multiplication: k

-les taux de fission du <sup>238</sup>U et du <sup>239</sup>Pu rapportés à la fission du <sup>235</sup>U:  $\sigma_{fU238}/\sigma_{fU235}$ ,  $\sigma_{fPu239}/\sigma_{fU235}$ 

-le rapport des coefficients de danger: DK U235/DKPu239

-le temps de vie:  $\ell$  .

Les taux de fission et les coefficients de danger sont ceux mesurés au centre des assemblages. Les 151 mesures sont choisies parmi 36 assemblages; en plus des 34 assemblages utilisés dans BARBAK (2. 1. 2), nous traitons RAPSODIE et sa maquette ZPR III-44.

Les paramètres théoriques comparés aux valeurs expérimentales sont calculés dans l'approximation de la diffusion en géométrie sphérique à 25 groupes pour des assemblages homogènes critiques avec réflecteur. En particulier, les coefficients de multiplication sont ceux déjà utilisés dans BARBAK.

<u>Sections efficaces</u>. Dans cette première étape, les variations affectent les sections efficaces dont l'influence a été jugée prépondérante dans l'étude précédente (section 1), soit:

<sup>235</sup>U: capture, fission et  $\nu$ 

<sup>238</sup>U: capture, fission, diffusion inélastique et  $\nu$ 

<sup>239</sup>Pu: capture, fission et  $\nu$ .

Nous faisons l'hypothèse que les facteurs d'autoprotection sont exacts. Les variations ne concernent <u>que les sections efficaces à dilution infinie</u> et sont limitées <u>aux quatre premiers groupes</u> d'un découpage en énergie à cinq grands groupes:

1.	0,821	-	14,5	MeV
2.	0,111	-	0,821	MeV
3.	9,12	-	111	ke V
4.	0,275	-	9,12	keV
5.	Therm.	-	275	eV

Pour chacun de ces quatre premiers groupes les coefficients  $\alpha$  (équation 1) sont calculés dans l'approximation de la diffusion à zéro dimension (code PERTUS) avec le découpage large précédent.

Les sections efficaces à cinq groupes sont obtenues par pondération sur le spectre au centre de chaque assemblage des sections efficaces à 25 groupes. Cette limitation du nombre de groupes où les variations peuvent avoir lieu est nécessaire puisque nous imposons un système surabondant. Ce premier ajustement porte sur 33 sections efficaces; 151 mesures sont utilisées.

<u>Poids</u>. Actuellement, nous supposons que l'erreur x sur un paramètre mesuré est constante quel que soit l'assemblage:

 $\begin{array}{rll} k & : & x = \pm \ 1\% \\ \\ \frac{\sigma_{f \ U238}}{\sigma_{f \ U235}} & : & x = \pm \ 2,5\% \\ \\ \frac{\sigma_{f \ Pu239}}{\sigma_{f \ U235}} & : & x = \pm \ 1\% \end{array}$ 

 $\begin{array}{l} \ell \ : \ x = \pm \ 10\% \\ \\ \frac{\rm DK \ U235}{\rm DK \ Pu239} \end{array} : \ x = \pm \ 2\% \end{array}$ 

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Jusqu'à présent, nous n'avons fait varier que le coefficient K(p) (équation 3) correspondant au coefficient de multiplication, de telle manière que le pourcentage dû à ce paramètre dans la somme initiale  $E_1^2$  des carrés des résidus (équation 2) varie de 20 à 75%.

Les résultats que nous allons présenter, au niveau actuel de l'étude, correspondent au pourcentage de 47,5% dû au coefficient de multiplication dans  $E_i^2$ .





<u>Résultats préliminaires</u>. L'ensemble des variations des sections efficaces obtenues est donné dans le tableau IV. La figure 2 met en évidence le gain apporté par l'ajustement sur les coefficients de multiplication calculés.

La comparaison des écarts-types et des valeurs moyennes avant et après ajustement, soit sur l'ensemble des mesures, soit pour chaque type de paramètre, permet de constater l'amélioration apportée au jeu qui serait ainsi modifié (tableau V).

# 2.2.3. Validité des résultats

a) Les variations obtenues des sections efficaces et les valeurs ajustées des paramètres sont pratiquement indépendantes du poids donné au coefficient de multiplication dans une gamme telle que le pourcentage dû à ce paramètre dans la somme initiale des carrés des résidus (équation 2) varie entre 35% et 65%.

	· · · · ·		Variations (%)				
Réaction	Energie	235 U	239Pu	238 U			
·	0,821 - 14,5 MeV	20	-80	-15			
	0,111 - 0,821 MeV	17	<b>4</b> 5 ′	- 4,8			
Capture	9, 12 - 111 keV	. 20	- 3	-15			
	0, 275 - 9, 12 keV	-30	60	- 4, 9			
	∫ 0,821 - 14,5 MeV	6	- 7	- 6			
	0,111 - 0,821 MeV	-4	8,3				
Fission	9,12 - 111 keV	-6	-18	·			
	0,275 - 9,12 keV	-2, 5	15,5				
	0,821 - 14,5 MeV	+ 4, 3	5,2	+ <b>4</b>			
,	0,111 - 0,821 MeV	- 3,8	-3				
U	9,12 - 111 keV	- 0,1	-2				
	0,275 - 9,12 keV	-2,0	-2				
	·						
Diffusion inélastique <sup>238</sup> U	$\frac{d\sigma}{d\sigma}(1 \rightarrow 2) = -22\%$ $\frac{d\sigma}{d\sigma}(1 \rightarrow 2)$	→3) = -23%	$\frac{d\sigma}{\sigma}$ (2 $\rightarrow$ 3) = +1	76			

# TABLEAU IV. VARIATIONS DES SECTIONS EFFICACES OBTENUES PAR LA METHODE BARRAKA

b) Considérons pour chacun des trois éléments étudiés l'ensemble des variations des sections efficaces retenues (tableau IV). Nous avons vérifié, en particulier sur l'assemblage MASURCA 1-A, que, d'après ces variations, pour le jeu Cadarache, le <sup>239</sup>Pu et le <sup>235</sup>U sont surréactifs, et le <sup>238</sup>U est sous-réactif. Ces résultats sont en parfait accord avec ceux de la méthode BARBAK (2.1.2). Qualitativement, ceci est évident pour le <sup>235</sup>U: dans le domaine d'énergie où le flux neutronique est maximal pour un grand réacteur rapide (10 keV - 1 MeV), la méthode propose d'augmenter la capture et de diminuer la fission et le paramètre  $\nu$  de cet élément.

c) L'écart-type final du coefficient de multiplication ajusté donné par la version actuelle de BARRAKA, soit 0,7%, doit se comparer à l'écart-

# TABLEAU V. MOYENNES ET ECARTS-TYPES AVANT ET APRES AJUSTEMENT

Paramètre	Nombre de mesures	Moyenne initiale X <sup>c</sup> /X <sup>m</sup>	Moyenne finale xaj/xm	Ecart-type initial (%)	Ecart-type final (%)
k	35	1,012	1,000	2,0	0,7
Q	25	0, 980	0,951	8, 4	9, 7
of U238 of U235	33	0,967	0, 995	10,0	8,3
of Pu239 of U235	32	0,978	0, 999	3,4	2,5
<u>DK U23</u> 5 DK <sub>Pu239</sub>	26	1,039	1,005	6,7	4, 3
Total	151	0,994	0,992	6,6	5,7

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type final donné par BARBAK dans un ajustement portant uniquement sur les trois éléments <sup>235</sup>U, <sup>238</sup>U, <sup>239</sup>Pu, soit 1% (et non à la valeur 0,3% du tableau III). Augmenter le nombre d'éléments dont les sections efficaces sont ajustées dans BARRAKA, tout en conservant un système largement surabondant, permettra donc de diminuer l'écart-type final sur le coefficient de multiplication, mais aussi les écarts-types des autres paramètres ajustés.

d) Bien que ces résultats soient préliminaires (tableau IV), nous avons voulu situer par rapport aux mesures expérimentales les valeurs des sections efficaces modifiées par BARRAKA, <u>mais il ne s'agit que de</u> tendances:

Fission	$-\frac{235}{0}$ U		
	E > 1 MeV	:	Evaluation de Parker [17]
	10  keV < E < 1  MeV	:	Limites inférieures des points expérimentaux de White [18] et Knoll et Ponitz [19]
	E < 10  keV	:	Résultats de l'expérience
Fission	155100 - 200 U       E > 1 MeV       : Evaluation de Park         10 keV < E < 1 MeV	PEIREL [20].	

Plutôt que  $\sigma_f$  nous prendrons le rapport  $\sigma_{fPu239}/\sigma_{fU235},$  qui est mesuré le plus souvent.

E > 1 MeV	:	Mesures de Allen et Ferguson [21]
0,1 MeV < E < 1 MeV	:	Le rapport ajusté est toujours supérieur de 10% aux limites supérieures des points expéri- mentaux de White [18]
10  keV < E < 100  keV	:	Résultats de White [18].

#### Capture - <sup>235</sup>U

Seul le rapport  $\alpha = \sigma_c/\sigma_f$  est mesuré, comme pour le <sup>239</sup>Pu. La valeur ajustée de  $\alpha$  serait systématiquement supérieure de 10% aux mesures de Lottin et Weston [22] ou de Saussure [23].

#### Capture - <sup>239</sup>Pu

Le rapport  $\alpha$  ajusté est également supérieur de 15% aux mesures de Lottin et Weston [22], sans cependant atteindre les valeurs de Uttley et James [24] en dessous de 10 keV.

# Capture - <sup>238</sup> U

Mesures de Hanna et Rose [25], Lyon et Macklin [26], Belanova [27], Moxon et Chaffey [28] dans le domaine d'énergie compris entre 1 MeV et 20 keV.

## Paramètre v

E > 1 MeV

 Pour tous les éléments, le ν doit être augmenté; <sup>235</sup>U, <sup>238</sup>U; évaluation du BNL [29]; <sup>239</sup>Pu; mesures de Mather et al. [30]

E < 1 MeV

: Pour tous les éléments, la diminution proposée du  $\nu$  nous rapproche des mesures de Sowerby et al. [31] pour l'étalon <sup>252</sup>Cf.

e) Il n'est pas question actuellement de modifier les données de base, ni d'utiliser cette méthode comme moyen d'évaluation automatique. Si le sens de ces variations correspond à la réalité, leur amplitude n'est sans doute pas rigoureusement exacte: la limitation du nombre d'éléments peut entraîner un ajustement forcé sur une section efficace de l'un d'entre eux, alors que la source d'erreur provient d'un autre corps non ajusté (fer par exemple). Rappelons également que nous supposons initialement que les erreurs dues aux méthodes de calcul sont nettement inférieures aux erreurs dues aux incertitudes sur les données de base. Cependant, les éléments dont nous ajustons les sections efficaces dans la version actuelle sont ceux dont l'influence est prépondérante. Cette méthode nous permet dans l'immédiat, pour de nombreux paramètres mesurés dans-les assemblages critiques, d'améliorer la précision des prévisions par calcul.

## CONCLUSION

Les incertitudes actuelles sur les données de base sont telles que les paramètres d'un grand réacteur rapide ne peuvent être calculés avec une précision suffisante pour un projet. Nous souhaitons que les demandes de mesures de données fondamentales que nous avons pu formuler avec leur précision soient rapidement satisfaites.

L'utilisation des paramètres mesurés dans les expériences rapides critiques nous a permis d'améliorer la précision de plusieurs paramètres calculés à partir du jeu Cadarache:

-Sans modifier le jeu, la méthode BARBAK prédit le coefficient de multiplication d'un nouvel assemblage avec un écart-type de 0,3%;

-Au contraire, la méthode BARRAKA permet d'ajuster les données multigroupes les plus importantes du jeu initial, donc de mieux calculer, non seulement le coefficient de multiplication, mais aussi tous les paramètres d'un réacteur.

Cette seconde méthode, plus attirante pour le physicien car elle s'attaque à la source des erreurs, est actuellement développée à Cadarache. Les extensions prévues comprennent l'étude systématique des poids, l'ajustement des sections efficaces d'autres éléments (par exemple le fer), l'analyse de nouveaux paramètres et assemblages. Une amélioration importante pourrait être envisagée si nous disposions de résultats expérimentaux provenant d'un plus grand nombre d'assemblages au <sup>239</sup>Pu et de paramètres mesurés plus sensibles aux réactions de capture ( $\overline{\alpha} = \sigma_c / \sigma_f$ ).

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# DISCUSSION

J. L. ROWLANDS: In studying the extent to which a parameter like breeding is sensitive to cross-section changes, one can either allow the  $k_{eff}$  to vary or adjust the enrichment so as to maintain criticality. The second procedure is the more appropriate when arriving at the cross-section measurement requirements for reactor design. With regard to the first part of your paper, which method did you use?

J. Y. BARRE: Firstly, we were not studying the sensitivity of a parameter with respect to a cross-section, but rather the margin of error in a parameter arising from the estimated uncertainty in regard to a cross-section.

Secondly, the reactor has in point of fact to be brought back to criticality again after the initial perturbation. I would go so far as to say that these secondary perturbations, if I may so term them, have a major effect on the internal breeding rate; for instance, as regards the effect of uncertainties in the <sup>239</sup>Pu fission cross-section, the primary and secondary perturbation effects tend to cancel each other out.

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# THE DATA REQUIRED FOR FAST REACTOR SAFETY ASSESSMENT BY PROBABILITY METHODS

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#### Abstract

THE DATA REQUIRED FOR FAST REACTOR SAFETY ASSESSMENT BY PROBABILITY METHODS. Probability methods for the assessment of reactor safety have been developed in the UK to the point where it is possible to make some quantitative comparisons of the safety of differing reactor types measured against acceptable criteria for a given site. One particular method for rapid assessment of preliminary designs has been used to show those areas of fast reactor physics and kinetics where further work could materially assist the designer by allowing relaxation of the standards required of reactor shut-down and emergency systems without impairing reactor safety.

### 1. INTRODUCTION

A considerable amount of work has been devoted recently in the UK to developing probability methods for the assessment of reactor safety. They are based on associating the consequences of any given reactor or plant failure in terms of the radioactivity subsequently released, with the probability of such an event taking place. Two possible sets of criteria to be used in assessing reactor safety have been described by Farmer [1] and by Adams and Stone [2].

A simplified method of analysis has been developed by Atomic Power Constructions Ltd. (APC) and its application to Advanced Gas-cooled Reactors (AGRs), pressure-tube water reactors and sodium-cooled fast reactors has been outlined in Refs [3], [4] and [5] respectively.

One of the major contributions of the method is to provide a means of comparing the relative contributions to safety made by plant items such as protective systems, emergency heat-removal systems and containment systems. In addition the worth of increased accuracy in basic data can be assessed in terms of the effect on the accuracy of the prediction of events subsequent to a primary plant failure.

This paper seeks to establish for large sodium-cooled fast reactors those physical and engineering data where increased accuracy or reliability would make a significant contribution to reactor safety. It is shown how such contributions could conceivably have economic benefits by allowing reductions in reliability standards required of some plant items.

# 2. OUTLINE OF THE METHOD OF ANALYSIS

The APC method of analysis has been described by Cave and Holmes [3] and only a brief outline is presented here. The method involves writing down all conceivable fault sequences and examining in more detail those which could lead to large  $(10^3 - 10^7 \text{ Ci} \, ^{131}\text{I})$  fission-product releases. The probability of each stage of the sequence taking place is assessed and the total probability calculated. The result is compared with the target defined by the safety criteria described in the next section. If the target probability for a release of the magnitude envisaged is not achieved, the improvement required in the reliability of the shut-down system, emergency system, containment, etc., is apparent. The simplicity of the method, which entails only simple hand calculations, also makes it easy to examine the effects of large perturbations in the original assumptions. The method is described in more detail in Appendix I.

# 3. CHOICE OF NUMERICAL SAFETY CRITERIA

In the absence of firm criteria to be used for licensing procedures in the UK and to provide a comparison of fast reactors with AGR, the present paper uses the criteria developed in Ref.[3]. These follow Farmer's proposals [1] as far as large releases are concerned and are based on a consideration of the effects of a single reactor during a 30-year life-time and do not take into account the complexities of a national reactor programme. Again it has been considered that if the probability attributed to any single primary fault does not exceed 1/10 of the acceptable total probability for the corresponding iodine-131 release, then summing over all the faults does not violate the acceptable total probability criterion.

At the lower end of the release spectrum, an economic criterion has been imposed such that the probability of individual primary faults leading to releases of 1000 Ci of iodine-131 should not exceed  $10^{-4}$  per reactor year or  $3 \times 10^{-3}$  in the reactor life. This criterion is based on a simple evaluation of the commercial risk to the operator arising from the prolonged outage which would be associated with faults leading to releases of less than 1000 Ci <sup>131</sup>I.

The proposed criteria for this paper are presented in Fig.1.

# 4. SALIENT FEATURES OF THE 1000 MW(e) CIVIL FAST REACTOR (CFR)

The salient features of the 1000 ME(e) CFR are described in Appendix II.

# 5. QUALITATIVE ANALYSIS OF CONCEIVABLE FAULTS IN CFR

A paper to the International Conference on the Safety of Fast Breeder Reactors [8] has described, in some detail, possible accident mechanisms which could lead to the release of significant quantities of radioactive fission products and plutonium from a fast reactor such as the Prototype Fast Reactor (PFR) or the CFR developed from it. Some of these mechanisms SM-101/50

are shown in Fig.2. A significant release can occur only if the primary containment is breached in some way, since the reactor vault roof is designed to contain the active gas blanket appropriate to the use of vented fuel in normal operation. The breach could occur in three ways:

(i) Lifting or cracking of the vault roof following an explosion or the formation of high pressure within the vault

(ii) Melting through the reactor tank and leak jacket by molten fuel

(iii) Failure of the gas blanket clean-up plant components or pipework.



FIG.1. Proposed safety and economic criteria.

It should be noted that the secondary containment would normally control releases through breach (iii) above and also small leakages under (i). Therefore a serious release can occur only if widespread fuel melting takes place or if sodium boiling produces a high enough pressure in the vault to produce significant leakage through the vault roof from an active gas blanket. This itself is unlikely unless accompanied by fuel melting.

Widespread fuel melting in the reactor core would occur only in one of the following sets of circumstances:

(a) The reactor fails to shut down prompty in the event of a severe primary fault, such as sub-assembly blockage, fuel pin failure, or loss of coolant flow.

(b) The reactor shuts down correctly after a primary fault but the removal of fission product decay heat is inadequate.





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Melting of one sub-assembly could also occur due to errors in the fuel handling procedures after the sub-assembly leaves the primary sodium.

# 6. THE UPPER LIMITS TO FISSION PRODUCT RELEASES IN FAULT CONDITIONS

Iodine has a strong affinity for sodium at reactor temperatures. Experiments involving injecting iodine or sodium iodide into sodium have given somewhat variable results, but it would appear that a decontamination factor (DF) of at least 1000 can be justified where the escaping fission products pass through a significant depth of liquid sodium. Some French work [9] has shown that even when the escaping iodine from molten fuel is accompanied by sodium vapour, the DF in passing through liquid sodium was as much as 100 000. For the purpose of this report a DF of 1000 has been assumed for iodine released under liquid sodium, including iodine released as methyl iodide. It has been further assumed that when sodium burns in air 10% of its iodine content is released.

The total inventories of fission products for a 1000 ME(e) CFR in continuous operation with unvented fuel are approximately:

Total iodine	$5 \times 10^8$ Ci
<sup>131</sup> I	$6 \times 10^7$ Ci
<sup>137</sup> Cs	$8 \times 10^4$ Ci
Rare gases (Kr and Xe)	$9 \times 10^8$ Ci

The CFR core as designed also contains some 2400 kg of plutonium in the form of plutonium oxide.

The upper limit to iodine releases is, however, rather less than the figures quoted above since it is difficult to see how the iodine could be released without travelling through at least some sodium or sodium vapour, and even if the sodium burned later on contact with the atmosphere, only about 10% of the available iodine would be released, say  $6 \times 10^6$  Ci <sup>131</sup>I. This release could only occur if the reactor failed to shut down correctly and the resulting nuclear excursion had sufficient energy to disrupt the primary containment to the point where the secondary containment was also breached by missiles or the pressure generated by burning sodium. It is also conceivable that, if the reactor were shut down but fission product heat removal were inadequate, fuel melting could lead to a critical assembly and a chain of events similar to that described above.

The upper limit release of rare gases has been taken as the total inventory  $(9 \times 10^8 \text{ Ci})$ , although there is evidence of significant retention in both fuel and sodium under some conditions. Within one or two kilometres of the site this would be equivalent, in terms of whole-body effects, to some  $10^7 \text{ Ci}$  <sup>131</sup>I; at greater distances its relative importance diminishes rapidly.

The possible upper limit release of plutonium is a little uncertain but has been pessimistically assumed to be 10% or 240 kg (which is equivalent in the more important radio toxic effects to about  $2.4 \times 10^6$  Ci <sup>131</sup>I [3]. Thus taking into account the pessimistic nature of the APC method [3], it is sufficient to evaluate the hazards in terms of the iodine release alone; taking  $10^7$  Ci <sup>131</sup>I as equivalent to the release of  $6 \times 10^6$  Ci <sup>131</sup>I and the shorter-lived iodines.

# 7. FREQUENCY OF PRIMARY AND SEQUENTIAL FAULTS

Those primary faults which could lead to upper limit releases are summarized in Table I together with some estimates of the possible sequential faults. The frequencies quoted have been estimated by reference to a wide range of sources and are generally in close agreement with those previously quoted in Ref.[3].

It will be seen that there are several chains of events which could conceivably lead to a release of about  $6 \times 10^6$  Ci <sup>131</sup>I. All these chains involve postulating an energy release in the reactor vault roof leading to breaching of the secondary containment. They derive from failure of the reactor to shut down on demand following flow failure or from heat removal failure in the core, rather than from failure of structural components, which is much less likely to occur.

Detailed calculations [10] have shown that an energy release of the magnitude required to collapse the PFR vault roof is most unlikely. However, some small finite possibility of its occurrence does exist and this has been put at  $10^{-3}$  per incident in Ref.[8]. It is expected that similar probabilities would pertain to the chance of exceeding design limits for the CFR vault roof.

Since spent reactor fuel is handled only one sub-assembly at a time, the maximum release in a fuel handling accident is some  $1.5 \times 10^4$  Ci  $^{131}$ I for a 30-day cooled sub-assembly.

# 8. PHYSICS DATA REQUIRED FOR PREDICTION OF REACTOR BEHAVIOUR UNDER FAULT CONDITIONS

It will be seen from Table I that with the exception of fuel handling accidents, major releases from a CFR of the deisgn considered would always be associated with a nuclear explosion of some magnitude. The probability ascribed to such an explosion therefore always features as a part of the overall probability of large-scale fission-product releases.

For the purposes of Table I a simple distinction has been made between what are termed 'small' and 'large' explosions based on the ability of the primary and secondary containments to fulfil their functions afterwards. In the absence of detailed information values of  $10^{-1}$  and  $10^{-4}$ have been assigned to the possibility of a 'small' or 'large' explosion respectively. These are the probabilities of such an event occurring per occasion on which all or most of the fuel melts.

# TABLE I. PROBABILITIES FOR ACTIVITY RELEASES TO ATMOSPHERE (1000 MW(e) CFR)

Primary fault	Frequency (per reactor life)	Secondary fault	Frequency (per primary fault)	Subsequent events	Frequency	Overall probability	Release (Ci <sup>131</sup> ])
1. Loss of coolant faults Self-failure of primary tank	10 <sup>-5</sup>	Failure of leak jacket	10-5			10 <sup>-10</sup>	104
2. Loss of coolant flow faults							
Loss of power to primary pumps	10	Failure to trip	10-6	<ul> <li>(a) Small explosion</li> <li>(b) Large explosion causing failure of primary and secondary containment</li> </ul>	10 <sup>-1</sup> 10 <sup>-4</sup>	10 <sup>-6</sup> 10 <sup>-9</sup>	< 100 . 10 <sup>7</sup>
Failure of primary pump outlet duct	10-4	Failure to trip	10-6	(a) (b)	10 <sup>-1</sup> 10 <sup>-4</sup>	10-11 10-14	<100 10 <sup>7</sup>
Sputious operation of flow controller Failure of diagrid causing flow failure	1 10 <sup>-5</sup>	Failure to trip Failure of rods to enter core	10-0	(a) (b) (a)	10 <sup>-1</sup> 10 <sup>-4</sup> 10 <sup>-1</sup>	10-7 10-10 10-7	<100 107 <100
				(b) .	10-4	10-10	107
3. <u>Reactivity faults</u> Rod withdrawal at power	1	Failure to trip	10-6	(a)	10-1	10-7	< 100
Rod withdrawal during start-up	10-2	Failure to trip	10-6	(b) (a) (b)	10 <sup>-4</sup> 10 <sup>-1</sup> 10 <sup>-4</sup>	10-10 10-9 10-12	10" <100 10"
Rod withdrawal during shut-down (Operator error)	10-3	Failure to trip	10-6	(a) (b)	10 <sup>-1</sup> 10 <sup>-4</sup>	10 <sup>-10</sup> 10 <sup>-13</sup>	<100 10 <sup>7</sup>
4. Loss of secondary coolant faults							
Loss of power to secondary pumps (Operator error)	10-3	Failure to trip	10-5	(a) (b)	10 <sup>-1</sup> 10 <sup>-4</sup>	10 <sup>-10</sup> 10 <sup>-13</sup>	<100 10 <sup>7</sup>
Turbine trip	100	Failure to trip	10-6	(a) (b)	10 <sup>-1</sup> 10 <sup>-4</sup>	10 <sup>-5</sup> 10 <sup>-8</sup>	<100 107
Secondary sodium fire	10-3	Affecting all circuits	10-5*			10-8	107
Spurious operation of flow controller	1	Failure to trip	10-6	(a) (b)	10 <sup>-1</sup> 10 <sup>-4</sup>	10-7 10 <sup>-10</sup>	<100 10 <sup>7</sup>

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TABLE I (cont.)

		·····					
Primary fault	Frequency (per reactor life)	Secondary fault	Frequency (per primary fault)	Subsequent events	Frequency	Overall probability	Release (Ci <sup>131</sup> 1)
5. Failure of emergency heat removal systems							
Reactor trip for reasons in 1-4	100	Failure to supply cooling within a few hours	10-6	(a) (b)	10 <sup>-1</sup> 10 <sup>-4</sup>	10 <sup>-5</sup> 10 <sup>-8</sup>	<100 10 <sup>7</sup>
Spurious reactor trip	100	Failure to supply cooling within a few hours	10-6	(a) (b)	10-1 10 <sup>-4</sup>	10 <sup>-5</sup> 10 <sup>-8</sup>	<100 10 <sup>7</sup>
Turbine disintegrates	10-3	Debris causes loss of all secondary sodium systems	10-5**			10-8	10 <sup>7</sup>
6. Fuel handling faults							
Fuel handling accident outside the reactor vessel involving burning	10-4	Containment intact clean up plant working	1			10-4	<100
01 1 5/A		Containment open	10-2			10-6	2×104
6. Fuel handling faults Fuel handling accident outside the reactor vessel involving burning of 1 S/A	10-4	secondary sodium systems Containment intact clean up plant working Containment open	1 10 <sup>-2</sup>			10 <sup>-4</sup> 10 <sup>-6</sup>	<100 2×104

\* This figure could be achieved by careful attention to secondary sodium pipework layout and the overall probability for this accident could be greatly reduced by containing the system in an inert atmosphere.

\*\* This figure could readily be achieved by siting the steam generators away from the turbine, by placing the turbine end-on to the generators and by providing blast walls.

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Another requirement for many of the fault conditions leading to major activity releases is failure of the reactor to trip. By this is meant failure of the shut-down system to inject sufficient negative reactivity in time to prevent sodium boiling and/or widespread fuel failure.

The third factor which must be considered in relation to large activity releases is the reliability of the post incident heat removal systems, since failure of these systems to operate on demand could lead to fuel failure and the possibility of a nuclear explosion.

Of these three aspects of reactor technology the last is largely a question of careful engineering, layout and provision of adequate equipment to give the required redundancy.

Reactor physics plays a more important role in the calculation of the appropriate reliability figures for the other two aspects. For example, Hicks [10] and others have shown that the magnitude of any possible nuclear explosion is highly dependent on the value of the Doppler coefficient of reactivity and to a lesser extent on the parameters involved in the equation of state. The time available to trip the reactor for various fault conditions also depends on the Doppler coefficient, and on the factors affecting the formation of superheat conditions in the sodium.

In addition there are many other non-nuclear physical properties and engineering considerations for the core components which influence the course of major core incidents. However, most of these are now known with sufficient accuracy or are parameters which are not in general amenable to alteration for safety reasons.

# 9. RELIABILITY OF REACTOR SHUT-DOWN AND EMERGENCY SYSTEMS

In Ref. [5] it has been shown that for a 1000 MW(e) civil fast reactor suitable for urban siting the failure rates typically required of the reactor shut-down and emergency systems are  $10^{-6}$  and  $5 \times 10^{-7}$  per demand, respectively. These figures are calculated on the assumption that in the event of a core melt-down there is a 1 in 10 chance of a prompt critical assembly occurring and a further 1 in 1000 chance that such an assembly would lead to an explosion of such a magnitude that both primary and secondary containments were breached.

Obviously if these figures could be reduced (i.e. the events made less likely), the table shows that the performance standards required of the shut-down and emergency systems could be relaxed. For example, for any given design the vault and roof may be designed to withstand the effects of a certain size of explosion, typically they might be calculated using the most pessimistic estimate currently available for the Doppler coefficient. This size of explosion could then be taken as the dividing line between 'small' and 'large' explosions. Further refinement of the calculations or experimental evidence might show that the chance of explosion exceeding the dividing line was 100 times less likely than previously thought. This would enable the cost of the double shut-down system to be saved as the required reliability would now be 1 failure per 10<sup>4</sup> demands.

#### 10. THE DATA REQUIRED

To carry out a complete analysis for a fast reactor of the type outlined above and in Ref.[5], it will be necessary to assemble a considerable volume of information.

Some of this data falls in the category of plant performance and reliability and some, of more interest to this Symposium, in the field of reactor physics. Most of this data is not available at the present time. The engineering data could probably be assembled from the records of manufacturers, electricity supply companies, etc., but the physics data has yet to be produced.

It should be noted that what is required is not merely the maximum expected size of an explosion, but the chance of the maximum expected size being exceeded. Similarly, considerably more information is needed to assess the reliability required from the reactor shut-down system, and the prospects of obtaining this from any proposed design. The way in which the progress of the fault affects the quickness of response of the sensing devices is particularly important in this context. Thus the physics data required can be listed and the cost of obtaining it for any given reactor design can be weighed against the cost of providing more reliable shutdown systems or stronger containments.

Taking a factor of 10 in probability as a minimum standard of accuracy, we can see that some data are already available with sufficient accuracy. Doppler coefficient, for example, and the equations of state for  $UO_2$  are probably known to this accuracy, but the rate of reactivity addition for various types of fuel melt-down for any given reactor design is not nearly so well known. Other areas where further accuracy is required are the question of the relevance of the simple geometries and models used in the calculations to the complex core geometries of real reactors.

To assess trip reliability considerably more data than at present exist are required to trace the course of sub-assembly incidents and to show that for all possible types and degree of faults there exist sensing devices with sufficient speed of operation and sensitivity to detect the faults in time. In particular the factors affecting superheating of sodium under reactor conditions and the effects of sodium coefficient require close attention. In this connection the sodium coefficient is now known with more than sufficient accuracy.

#### 11. CONCLUSIONS

Reference [5] used a simple method of probability analysis to demonstrate the suitability of a large sodium-cooled fast reactor for urban siting. This paper has extended these ideas to show that increased accuracy in certain areas of basic physics data could lead to a relaxation of the requirements for some reactor safety systems laid down in Ref.[5]. It remains for the individual designer to weigh the relative cost of obtaining this accuracy against the saving in plant costs, for his particular design.

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#### APPENDIX I

# MORE DETAILS OF THE APC METHOD

#### A.1. INTRODUCTION

The object of this Appendix is to set down in more detail certain aspects of the APC method of probability analysis. The following topics are considered:

Method of summation of risks

The pessimistic nature of the APC method.

# A.2. METHOD FOR SUMMATION OF RISKS

This is most conveniently illustrated initially by consideration of a criterion based on economic considerations, thus:

Let  $P_i$  denote the probability of some fault i which leads to an outage during the life of the reactor. Denote the cost of the enforced outage due to fault i by  $\pounds C_i$ . Then the operator's expectation of loss due to this fault is  $\pounds p_i C_i$  and clearly his total expectation of loss due to all primary faults which lead to outages during the life of the reactor is  $\pounds \Sigma p_i C_i$ .

Similarly, the 'expectation of damage to the public' due to faults which lead to the release to atmosphere of fission products can be denoted by  $\Sigma p_i R_i k_i$  where  $R_i$  denotes the release, which may, in general, be taken as the <sup>131</sup>I release, and  $k_i$  is a factor relating release, in curies of <sup>131</sup>I, to number of casualties. If a linear relation between release and casualties is assumed,  $k_i$  is a constant.

In the course of the analysis the probabilities of the various faults are estimated, together with the corresponding releases of <sup>131</sup>I. In establishing the safety criteria it is convenient, in practice, to specify the maximum acceptable probability corresponding to some fixed value of <sup>131</sup>I release, e.g. Farmer [1] has suggested that the probability of a release of 10<sup>6</sup> Ci <sup>131</sup>I should not exceed 1 per 10<sup>7</sup> reactor years, making the assumption that only one such release need be considered. Thus to establish whether the risk, summed over all faults, is acceptable it is permissible to compute  $\Sigma(p_i C_i k_k / C_r k_r)$  where  $C_r$  is some specific size of release used for reference purposes, in order to arrive at a mean probability to compare with the proposed criterion. In the APC method it is convenient to use the practical upper limit for the <sup>131</sup>I release for reference purposes.

# A.3. THE PESSIMISTIC NATURE OF THE APC METHOD

The frequencies assumed for primary faults (see Table I of this paper) are best estimates rather than upper limits; they could therefore contain some degree of optimism. SM-101/50

The reliability assumed for the reactor shut-down system and emergency cooling system is treated pessimistically. It is assumed that the stated failure rate applies to operation of the system within the maximum time assumed in the design. In the event of failure to operate within this time it is quite likely that they would still operate in time to prevent core melt down. However, no credit is taken for this possibility.

Thus for each primary fault which necessitates operation of these systems it is assumed that either the reactor shuts down correctly, or the fault is uncorrected and leads to core melt down and the practical upper limit release.

In mathematical terms, in the event of a primary fault of probability  $p_i$  which necessitates shut-down, it is assumed that there is a probability of q of failure to shut down correctly and a probability (1-q) of shutting down correctly. Thus the probability of the release of  $1 \times 10^7$  curies of  $1^{31}$ I is  $qp_i$ , and the probability of zero release is  $(1-q)p_i$ . In some instances two alternative paths following a primary fault can readily be identified which may lead to different shut-down requirements. The two paths, which must have a combined probability of unit, are each treated in the way described above.

#### APPENDIX II

#### SALIENT FEATURES OF THE 1000 MW(e) CFR DESIGN

The design chosen for this paper is the 1000 MW(e) CFR described by representatives of the three British Consortia at the BNES Conference in May 1966 [12]. It is a direct development of the Prototype Fast Reactor being built at Dounreay for the UKAEA which has been fully described in Refs [6, 7 and 13]. Brief details only are given below of the CFR design, which differs essentially from PFR in being four times as large in output, and in that PFR is situated on an isolated site.

#### 1. CORE DESIGN

The CFR core consists of 295 hexagonal sub-assemblies each comprising 325 stainless steel clad, mixed uranium/plutonium oxide fuel pins. The pins are supported by grids within a stainless steel hexagonal wrapper. Eight tantalum rods are used for control and 10 more rods are used to maintain sub-criticality during fuel handling and to assist reactor shut-down.

# 2. PRIMARY CIRCUIT

The entire primary circuit is contained within a cylindrical primary tank of stainless steel, surrounded by a separate steel leak jacket. The leak jacket is contained within a reinforced concrete vault below ground level. Primary sodium is circulated through the core and 18 intermediate heat exchangers by 6 mechanical pumps in parallel. These pumps are totally immersed in the primary sodium and are driven by electric motors situated above the vault roof. The argon gas blanket covering the primary sodium is held at about 1  $lb/in^2$  above atmospheric pressure.

## 3. SECONDARY CIRCUIT AND STEAM GENERATORS

The secondary sodium circuit is in the form of 6 separate loops each connected to an intermediate heat exchanger. Each loop has its own circulatory pump and 3-stage steam generator. The steam generators are provided with bursting discs to relieve pressure and vent waste products in the event of a sodium/water reaction following a tube failure.

#### 4. FUEL HANDLING

Replacement of spent fuel in the core with fresh fuel from the rotary store within the reactor vessel is carried out with the reactor shutdown. Movement of fuel to and from the store and the fuel handling caves external to the reactor vault can, however, be carried out with the reactor on load.

The reactor vault is covered by a reinforced concrete roof fitted with a rotating shield concentric with the core. Refuelling entails rotating this shield and therefore requires that the control and shut-off rods are disconnected from their mechanisms and must therefore remain in the core throughout reactor refuelling operations. The charge machine can handle only one sub-assembly at a time and can only unlatch it in the lowered position. The sub-assembly is handled entirely immersed in sodium.

Spent fuel from the storage rotor is handled entirely in sodium-filled buckets which can be transferred one at a time in an argon atmosphere to the buffer store and hence to the argon-filled examination and decontamination caves.

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#### DISCUSSION

D. STEGEMANN: Would you indicate how you arrived at the figures quoted in the example you mentioned in your oral presentation, for primary failure consisting in loss of power to primary pumps an estimated frequency of 10 per reactor life-time and for associated secondary failure 10<sup>-4</sup>?

L. CAVE: The assumed incidence of primary pump failure is based on the experience of the United Kingdom electrical supply system, on the assumption that the primary pumps are driven by electric motors taking their supply from the grid via transformers. The data available suggest that the figure of 10 failures per reactor lifetime (i.e. once in 3 years) is reasonable. In practice it might vary from 1 to 30. Other figures might be appropriate for other arrangements of pump drives or electrical supplies.

As far as secondary failures are concerned, the incidence of failures on demand of reactor shut-down systems has been investigated closely in the United Kingdom for our Magnox and advanced gas-cooled reactors. It is considered that with a single shut-down system (based on two effective trip parameters for each fault condition and with "2 out of 3" logic in each of the trip channels) a failure rate of 1 in  $10^4$  demands is reasonable. For fast reactors the time available to detect some faults and to shut the reactor down is less. The adequacy of the trip sensor time-response must therefore be viewed with some reservation for the time being, and it would be desirable to have function data on, for example, sodium superheating. However, if function data are not forthcoming, it should be possible, by ingenuity in design, to devise alternative sensors with time constants which would be fully acceptable.

K.E.J. WIRTZ (Chairman): How does British industry feel about the wish of the Central Electricity Generating Board to have the safety report based on the probability analysis? The task might be formidable. L. CAVE: I agree that the preparation of a safety report based on probability analysis would involve considerable work. However, we have prepared such reports for recent AGR tenders, using the APC method of analysis described in the paper. I feel that the additional work has been well worth while because of the much greater appreciation one reaches of those features in the design which are most important to safety.
## SENSITIVITY STUDY OF FAST REACTORS USING GENERALIZED PERTURBATION TECHNIQUES

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#### Abstract

SENSITIVITY STUDY OF FAST REACTORS USING GENERALIZED PERTURBATION TECHNIQUES. A broad sensitivity study is presented relative to integral quantities of interest for fast reactor evaluation, namely sodium effects, Doppler coefficients, prompt neutron lifetimes, using generalized perturbation techniques. Deviations roughly typical of present uncertainties are assumed for differential nuclear data, and therefore the results obtained are fairly indicative of the inaccuracies to be associated with theoretical predictions.

#### 1. INTRODUCTION

The present work aims to analyse the influence of the different nuclear parameters on the assessment of integral quantities relevant to the reactor safety of typical fast ceramic reactor systems; namely, the sodium void coefficient, the Doppler coefficient and the prompt neutron life-time. For this purpose generalized perturbation techniques have been used which take into account the influence on the integral quantities consequent on changes of both the real and adjoint neutron flux. It is shown that the influence of changes of the adjoint flux on such quantities may be relevant in some cases. In this respect the present paper is an improvement on a previous paper [1], where only the effects relevant to real flux changes were considered.

The variations of the differential nuclear parameters correspond roughly to assumed uncertainties of data available at present and therefore the results obtained are fairly indicative of the inaccuracies to be associated with theoretical predictions. On the basis of this analysis it is then possible to indicate which data, at different energies, are most inaccurate and how much improvement in the integral quantities considered could be obtained by reducing some of the assumed uncertainties.

In the analytical study the multigroup diffusion theory has been adopted and the cross-section library by Abagyan [2].

#### 2. THE PERTURBATION METHOD ADOPTED

Let us consider a ratio  $\rho_{\text{bil}}$  of two bilinear functionals of the real and adjoint neutron fluxes  $\vec{\varphi}$  and  $\vec{\varphi}^*$ :

$$\rho_{\text{bil}} = \frac{\int \vec{\varphi} * \mathbf{H} \vec{\varphi} \, \mathrm{d} \vec{r}}{\int \vec{\varphi} * \mathbf{F} \vec{\varphi} \, \mathrm{d} \vec{r}} = \frac{\mathbf{Q}_1}{\mathbf{Q}_2} \tag{1}$$

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In particular  $\rho_{bil}$  may represent reactivity values and prompt neutron lifetimes. In the former case H represents a change  $\Delta P$  of the Boltzmann operator whereas in the latter case it represents the inverse diagonal matrix  $v^{-1}$ . F is the fission production matrix. If now a perturbation  $\delta P$  is introduced into the system without affecting the criticality,  $\rho_{bil}$  will generally change by an amount  $\delta \rho_{bil}$ . Using generalized perturbation methods [1, 3], the following equation may be written:

$$\frac{\delta \rho_{\rm bil}}{\rho_{\rm bil}} = \int \vec{\varphi} * \delta \mathbf{G} \, \vec{\varphi} \, \mathrm{d}\vec{\mathbf{r}} + \int \vec{\psi} * \delta \mathbf{P} \, \vec{\varphi} \, \mathrm{d}\vec{\mathbf{r}} + \int \vec{\varphi} * \delta \mathbf{P} \, \vec{\psi} \, \mathrm{d}\vec{\mathbf{r}}$$
(2)

where

$$\delta \mathbf{G} = \frac{\delta \mathbf{H}}{\mathbf{Q}_1} - \frac{\delta \mathbf{F}}{\mathbf{Q}_2} \tag{3}$$

and  $\vec{\psi} *$ ,  $\vec{\psi}$  are respectively the over-all importance and flux density functions corresponding to the ratio  $\rho_{bil}$  considered. The first term at the right hand side of Eq.(2) represents the change of  $\rho_{bil}$  due to direct effects, i.e. changes of the operator  $\Delta P$ , if any. The second and third terms represent indirect effects, the second being connected to real flux changes, the third to adjoint flux changes. The formulation of these integral terms seems to be equivalent to that which appears as the numerator of conventional perturbation expressions as represented by Eq.(1). Functions  $\vec{\psi} *$  and  $\vec{\psi}$  are given by the following equations:

 $\vec{\psi} = \sum_{i=1}^{1} \vec{\psi}_{i}$ 

$$\vec{\psi}^* = \sum_{i=1}^{I} \vec{\psi}_i^* \tag{4}$$

where  $ec{\psi}_{\mathbf{i}}^*$  and  $ec{\psi}_{\mathbf{i}}$  are solutions of the iterative systems

$$\begin{cases}
-A * \vec{\psi}_{1}^{*} = G * \vec{\phi} * \\
-A * \vec{\psi}_{i}^{*} = F * \vec{\psi}_{i-1}^{*} \quad (i = 2, 3...) \\
(6)$$

$$\begin{cases} -A \vec{\psi}_i = F \vec{\psi}_{i-1} & (i = 2, 3...) \end{cases}$$
(7)

Operator A (and the adjoint A\*) are related to the loss (capture and leakage) and down-scattering processes. Since functions  $\vec{\psi}_i^*$  and  $\vec{\psi}_i$  converge to zero for  $i \to \infty$ , I and I' are decided by the convergence criterion chosen. It should be noted that systems (6) and (7) correspond to iterative equations normally used for calculating the adjoint and real fluxes.

The CIAP ID code solves systems (6) and/or (7) in multigroup diffusion theory and one dimension. The GLOBPERT ID code calculates  $\delta \rho_{bil} / \rho_{bil}$  values.

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	Pancake ox	tide system	Carbide	e system
	Core	Blanket	Core	Blanket
R (cm)	-	-	102.95	148.67
H (cm)	63.56	45.72	-	-
N <sup>Pu-239</sup>	0.000851	0.000199	0.00075	0,000206
NU-238	0.004253	0.010008	0.006272	0.012142
N <sup>Fe</sup>	0.01134	0.01015	0.004718	0.007166
N <sup>Cr</sup>	0.00326	0.00292	0.001355	0.00206
N <sup>Ni</sup>	0.00152	0.00136	0.000032	0.00096
NNa	0.01012	0.00704	0.01463	0.00946
N <sup>Be</sup>	0.00462	-		
NO	0.014828	0.020416		
<sub>N</sub> C	-	-	0.007022	0.012348

# TABLE I. COMPOSITIONS (in nuclei/cm $^3 \times 10^{-24}$ ) AND DIMENSIONS OF THE TWO REFERENCE FAST SYSTEMS

#### 3. FAST REACTORS STUDIED

The reactors considered in the present analysis are two plutoniumfuelled sodium-cooled breeders. They have been chosen according to present trends in fast reactor design. The first system is a pancake oxide reactor, the second system consists of a carbide cylindrical reactor with an L/D ratio close to one. The calculations have been performed in slab geometry for the pancake case, using a radial buckling  $B_r^2 = 0.000164$ to allow for the finite radial size, whereas for the carbide system the spherical geometry has been adopted. Compositions and dimensions of the two reference systems are illustrated in Table I.

#### 4. SODIUM VOID EFFECT

The integral parameter  $\rho_{\rm bil}$  chosen to study the sodium void effect is the reactivity value associated with the removal of 10% vol. of sodium from the system. The sensitivity analysis consists in the evaluation of the sodium effect changes consequent on assigned percentage variations of the group nuclear constants of the component elements. For the oxide pancake system the net sodium void reactivity effect is  $-0.928 \times 10^{-3} \, \delta k/k$ whereas for the carbide system it is  $-2.459 \times 10^{-4} \, \delta k/k$ . The results of the calculations are shown in Table II. For the clarity of exposition the 26 groups of the Abayan library have been condensed into four, as shown in Table III. The interpretation of the results requires that four different

		System		Carl	oide			Ox	ide	
Material	Param. σ	Gr. δσ/σ	1	2	3	4	1	2	3	4
	ν	0.02	- 19.73	- 44.82	46.49	39.55	- 2.73	- 7.86	2.84	8.00
	σ <sub>f</sub>	0.03	- 20.25	- 54.19	45.16	35.20	- 2.79	- 8.85	2.06	8,45
239 Pu	σc	0.05	0,38	0.99	- 25,88	- 26.19	0.05	0.57	- 2.42	- 6.63
	$\sigma_{in}$	0.10	4.38	-	-	-	0.45	-	-	-
	σ <sub>el</sub>	0.10	0.20	1.82	1.19	0.15	0.03	0.21	0.11	0.02
	υ.	0.02	- 25.09	•	-	-	- 2.86		-	-
	σ <sub>f</sub>	0.03	- 22.41	-	-	-	- 2.63	-	-	-
238 U	σ <sub>c</sub>	0.05	7.11	27.73	- 75.33	- 28.94	0.69	5.58	- 2.57	- 5.63
	σ <sub>in</sub>	0.10	77.66	-		-	5.43		-	-
	σ <sub>el</sub>	0.10	- 3.71	0.27	9.82	1.90	- 0.56	- 1,18	0.76	0.29
	σ <sub>in</sub>	0.10	- 40.78	•	•.	-	1.62	-	-	-
Na	σ <sub>el</sub>	0.10	- 11.98	- 3.99	82.09	3.67	- 0.82	- 0.26	6.62	0,13
<u> </u>	σ <sub>c</sub>	0.10	0.45	1,15	- 3.34	- 1.23	0.14	0,50	- 0.64	- 0.64
Fe	$\sigma_{in}$	0.10	16,93	-	-	-	3.56	-	-	-
	σ <sub>el</sub>	0.10	0,81	6.81	6.93	2.17	0.11	1.26	1.22	0.66
С	σ <sub>el</sub>	0.10	6.65	45.05	15.89	2.03	-	-	-	-
Ó	σ <sub>el</sub>	0.10	-		-	-	1.02	5.51	0.85	0.41
Be	σ <sub>el</sub>	0.10	-	•	•	-	0.90	2.66	0.68	0.16

TABLE II. SENSITIVITY VALUES RELATIVE TO THE SODIUM VOID EFFECT (in percent  $\delta \rho_{bil} / \rho_{bil}$ )

Coarse groups			1					2					3		
Fine groups	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15
E <sub>1</sub> (keV)	6500	4000	2500	1400	800	400	200	100	46.5	21.5	10.0	4.65	2.15	1.00	0.465
<u> </u>	<b></b>										L		· •		
Coarse groups								4							
Fine groups	16	17	18		19	20	21		22	23	24	4	25	Т	
E <sub>1</sub> (keV)	0.215	0.100	0.046	50.	0215	0.010	0,0046	5	0,00215	0.0010	0.000	<b>)4</b> 65	0,000215	0.0000	0252

## TABLE III. FINE AND COARSE GROUP STRUCTURE

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		System		Carl	oide			Oxi	ide	
Material	Param. σ	Gr. δσ/σ	1	2	3	4	1	2	3	4
	ν	0.02	- 20.08	- 44.86	47.00	40.00	- 2.75	- 7.69	2.96	8.00
	σ <sub>f</sub>	0.03	- 20.63	- 48.39	48.27	35.62	- 2.80	- 8.02	2,64	8.38
<sup>239</sup> Pu	σ <sub>c</sub>	0.05	0.40	2.86	- 22.51	- 26.46	0.05	0.76	- 2.09	- 6.85
	$\sigma_{in}$	0.10	2.42	-	-	-	0.20	-	-	
	$\sigma_{e1}$	0.10	0.03	0.10	0.26	0.0	0.0	0.01	0.02	0.0
	ν	0.02	- 23.61	-	-	-	- 2.60	-	-	-
	$\sigma_{\rm f}$	0.03	- 21.67	-	-	-	- 2.42	-	-	-
<sup>238</sup> U	σ <sub>c</sub>	0.05	7.05	35.24	- 65.04	- 29.92	0.65	5.61	- 2,39	- 6.01
	$\sigma_{in}$	0.10	47.12	-	-	-	3.29	-	-	-
	$\sigma_{el}$	0.10	0.36	1.78	2.15	0.24	0.03	0.18	0.11	0.02
	$\sigma_{in}$	0.10	12.34		-	-	0,66	-	-	-
Na	$\sigma_{el}$	0.10	6.09	16.90	17.70	1.13	0.36	1.04	- 0.52	0.07
	σ <sub>c</sub>	0.10	- 0.47	1.43	- 3.00	- 1.27	0.13	0.55	- 0.59	- 0.67
Fe	σ <sub>in</sub>	0.10	10.23	-		-	1.83	-	-	-
	σ <sub>el</sub>	0.10	0.81	2.48	4.59	0.55	0.16	0.47	0.85	0.12
с	σ <sub>el</sub>	0.10	7.00	20.02	16.73	1,54	-	-	•	-
0	σel	0.10	-	-	-	-	1.01	3,12	1.53	0,19
Ве	σ <sub>el</sub>	0.10	-	-	-	-	0.32	0.58	1.37	0.16

# TABLE IV. SPECTRAL TERMS RELATIVE TO THE SODIUM VOID EFFECT CONSEQUENT ON ADJOINT FLUX VARIATIONS (in percent $\delta \rho_{bil} / \rho_{bil}$ )

		System		Cart	vide			Oxi	de	
Material	Param. O	Gr. δσ/σ	1	2	3	4	1	2	3	4
	ν	0.02	. 0.0	0.0	0.0	0,0.	0,0	0.0	0.0	0.0
	σ <sub>f</sub>	0.03	0.05	0.10	0.10	0.08	0.01	0.01	0.01	0.01
239 Pu	σ <sub>c</sub>	0.05	0.0	0.03	0.10	0.09	0.0	0.0	0.01	0.01
	$\sigma_{in}$	0.10	0.10	-	-	-	0.01	-		-
	$\sigma_{el}$	0.10	0.15	1.47	0,97	0.10	0,03	0.18	0.11	0.02
	ν	0.02	0.0	-	-	-	0.0	-	、 -	-
	σ <sub>f</sub>	0.03 .	0.28	-	-	-	- 0.04	-	-	-
<sup>238</sup> U	σ <sub>c</sub>	0.05	- 0.07	- 0.06	0.31	0.14	- 0.01	- 0.02	0.04	0.03
	$\sigma_{in}$	0.10	- 5.42	-	, -	-	- 0.76	-	-	-
	σ <sub>el</sub>	0.10	- 4.32	- 4.59	8.17	1.72	- 0.61	- 1.54	0.76	0.28
	σ <sub>in</sub>	0.10	- 0.01	-	-	-	- 0.02	-	• •	-
No	$\sigma_{in}^*$	0.10	- 61.89	-	-	-	0.30	-	-	-
INZ .	o <sub>el</sub>	0.10	- 0.01	6.86	7.52	1.28	- 0.06	0.25	0.27	0.13
	$\sigma_{e1}^{*}$	0.10	- 22.38	- 47.93	65.69	1.47	- 1.50	- 2.63	- 4.04	- 0.04
	σc	0.10	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Fe	$\sigma_{\rm in}$	0.10	- 1.02	-	-	- 1	- 0.13	-	-	) - '
	o <sub>el</sub>	0.10	- 1.20	- 0.44	3.58	1.74	- 0.22	0.05	1.00	0.60
C	σ <sub>el</sub>	0.10	- 5.17	4.98	2.80	-	-	-	-	-
0	σ <sub>el</sub>	0.10	-	-	-	-	- 0.94	- 1.14	0.67	0.32
Ве	σ <sub>el</sub>	0.10			-	-	0.19	0.24	0.26	0.07

TABLE V. LEAKAGE TERMS RELATIVE TO THE SODIUM VOID EFFECT (in percent  $\delta \rho_{bil} / \rho_{bil}$ )

\* Direct effect

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		System		Carb	ide	,		Oxi	de	
Material	Param. σ	Gr. δσ/σ	1	2	3	4	1	2	3	4
	ν	0.02	- 0.44	- 1.47	0,19	1.06	- 0.47	- 1.53	- 0.52	3.50
	$\sigma_{\rm f}$	0.03	- 0.45	- 1.96	- 0.42	0.93	- 0.65	- 1.79	- 1.00	3.37
<sup>239</sup> Pu	σ <sub>c</sub>	0.05	0.01	0.03	- 0.65	- 0.65	0.0	0.12	- 0.18	- 0.58
	$\sigma_{in}$	0.10	0.15	-	-	-	0.09	-	-	
	σ <sub>el</sub>	0.10	- 0.01	0.0	0.01	0.0	- 0.01	0.0	0.0	0.0
	ν	0.02	- 0.51	-	-	-	- 0.36	-	-	-
	$\sigma_{\rm f}$	0.03	- 0.44	-	-	-	- 0.34	-	-	-
238 <sub>U</sub>	σc	0.05	0.12	0.10	1.03	- 0.15	0.08	0.42	2.08	0.33
	$\sigma_{in}$	0.10	2.14	-	-	-	0.90	-	-	-
	σ <sub>el</sub>	0.10	- 0.12	0.09	0.32	0.01	- 0.11	- 0.22	0.01	- 0.02
	σ <sub>in</sub>	0.10	0.51	-	•	-	0.18	-	-	-
Na	σ <sub>el</sub>	0.10	0.08	3.07	1.90	- 0.16	0.0	0.94	0.61	- 0.15
	σ <sub>c</sub>	0.10	0.01	0.01	- 0.14	- 0.06	0.02	0.06	- 0.15	- 0.16
Fe	σ <sub>in</sub>	0.10	0.43	-	-	-	0.56	-	-	-
	σ <sub>el</sub>	0,10	0.04	0.51	0.19	- 0.06	- 0.08	0.34	0.14	- 0.25
с	σ <sub>el</sub>	0.10	0.14	3.32	1.32	- 0.27	-	-	-	
0	σ <sub>el</sub>	0.10	-	-		-	0.07	2.21	0.67	- 0.44
Be	σ <sub>el</sub>	0.10	-	-	-	-	0.10	1.25	0.30	- 0.30

TABLE VI. SENSITIVITY VALUES RELATIVE TO DOPPLER EFFECT (in percent  $\delta \rho_{bil} / \rho_{bil}$ )

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effects should be distinguished, corresponding to the influence of the real and adjoint flux variations on the spectral and leakage terms into which the perturbation  $\delta P$  of Eq.(2) may be subdivided. In general it should be noted that there is a large contribution to the spectral terms due to the adjoint flux variation consequent on changes of the fission and capture properties of the fissile and fertile materials, as shown in Table IV. The inelastic and elastic scattering effect on the adjoint flux, and consequently on the spectral term, is comparable with that due to the real flux variation. As far as the leakage terms are concerned, their values are much smaller than the spectral ones, as may be seen in Table V, where also the direct effects of sodium cross-section changes, corresponding to the first term on the right-hand side of Eq.(2), are included. The variation of the real and adjoint fluxes due to the elastic scattering changes is also important in this connection.

It is interesting to notice that the direct effects consequent on sodium cross-section changes are of the same order of magnitude as many indirect changes consequent on flux variations. This shows in particular that when computing the sodium void effect one should take into consideration the rigorous formulation of the reactivity

$$\frac{\delta k}{k} = \frac{\int \vec{\varphi} * \Delta P \, \vec{\varphi}' \, d\vec{r}}{\int \vec{\varphi} * F \, \vec{\varphi}' \, d\vec{r}}$$
(8)

where  $\vec{\varphi}^{\dagger}$  represents the real flux after the sodium is removed. This may be evaluated by a good approximation using the generalized perturbation techniques adopted here [3].

#### 5. DOPPLER EFFECT

As far as the Doppler effect is concerned, the reactivity associated with a temperature rise in the core from 900 to 2100°K has been considered. For the oxide pancake system the value has been calculated as  $-1.547 \times 10^{-2} \delta k/k$ , and for the carbide system  $-1.391 \times 10^{-2} \delta k/k$ . The results of the calculations are shown in Table VI. It is interesting to note the role of the adjoint flux variation caused by changes of the  $\nu$  and  $\sigma_f$  of plutonium. This is shown in Table VII where also the direct contributions to allow a more detailed analysis are indicated . As expected, the adjoint flux variations consequent on changes of the  $\sigma_c$  of <sup>238</sup>U show an opposite trend. Real flux changes relevant to the Doppler effects are attributable particularly to the change in  $\sigma_c$  of <sup>238</sup>U in the third coarse group, and in general to the change in  $\sigma_{el}$  and  $\sigma_{in}$ , respectively, in the first and in the second and third groups. This is expected, as these cross-sections are directly involved in the softness of the spectrum to which the Doppler effect is related. Also here it seems of relevance that, with the rough uncertainties assumed, the main cause of the inaccuracy of Doppler effect evaluations appears to be cross-sections that are different from those involved directly in the Doppler effect itself.

#### 6. PROMPT NEUTRON LIFETIME

The prompt neutron lifetime for the pancake oxide and for the carbide system has been calculated as  $0.794 \times 10^{-6}$  s and  $0.571 \times 10^{-6}$  s. The

		System		Car	bide			Ox	ide	
Material	Param. o	Gr.	1	2	3	4	1	2	3	4
	ν	0.02	- 0.41	- 1,37	0.58	1,57	- 0.44	- 1,45	- 0.06	2.02
	σ <sub>f</sub>	0.03	- 0.42	- 1,49	0,58	1.45	- 0.44	- 1.50	- 0.09	1.92
	σ <b>*</b> f	0.03	-	-	- 0.37	- 0.45	-	-	- 0.41	1.48
239 <sub>Pu</sub>	σ <sub>c</sub>	0.05	0.01	0.15	- 0.33	- 1.02	0.0	0.19	- 0.04	- 1.52
	σ <b>*</b>	0.05	-		0.22	0.44	-	-	0.27	0,94
	$\sigma_{in}$	0.10	0.04	-	-	-	0.03	-	-	-
	$\sigma_{e1}$	0.10	- 0.01	- 0.03	0.0	0.0	- 0.01	- 0.03	- 0.01	0.0
	ν	0.02	- 0.49	-	-	-	- 0.36	-	-	-
	σ <sub>f</sub>	0.03	- 0.45	-	-	-	- 0.35	-	-	-
	σ <b>*</b>	0.03	0.0	-	-	-	-	-	-	-
238 U	σ <sub>c</sub>	0,05	0,12	1.11	- 0.96	- 1.28	0.08	0.93	- 0.07	- 1.54
	σ <b>*</b>	0.05	-	-	4.48	1.27	-	-	3.81	1,97
	$\sigma_{in}$	0.10	0.71	-	-	-	0.26	-	-	-
	o <sub>el</sub>	0.10	- 0.10	- 0.42	0.03	0.04	- 0.09	- 0.41	- 0.09	0.02
	σ <sub>in</sub>	0.10	0.18	•	-	-	0.05	•	-	-
Na	σ <sub>el</sub>	0.10	- 0.05	- 0.12	0.26	0.06	- 0.05	0,16	- 0.08	0.02
	σ <sub>c</sub>	0.10	0.01	0,04	- 0.05	- 0.05	0.02	0,10	- 0.02	- 0.16
Fe	$\sigma_{in}$	0.10	0.15	-	-	-	0,18	-	-	-
	o <sub>e1</sub>	0.10	0.02	- 0.11	0.10	0.05	- 0.09	- 0.23	- 0.01	0.04
С	σ <sub>el</sub>	0.10	- 0.01	0.0	0.31	0.07	-	-	-	-
0	o <sub>el</sub>	0.10	-	-	-	-	- 0.10	- 0,19	0.03	0.04
Be	<sup>o</sup> el	0.10	-	-	-	-	0.02	0.0	0.01	0.01

TABLE VII. SPECTRAL TERMS RELATIVE TO DOPPLER EFFECT CONSEQUENT ON ADJOINT FLUX VARIATIONS (in percent  $\delta \rho_{bil} / \rho_{bil}$ )

\* Direct effect

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		System		Car	bide			Oxi	de	
Material	Param. σ	Gr. δσ/σ	1	2	3	4	1	2	3	4
	ν	0.02	- 0,26	- 0,59	0,50	0.66	- 0.24	- 0.65	0.19	0.80
	σ <sub>f</sub>	0.03	- 0.27	- 0.90	0.09	0.44	- 0.25	- 0.86	- 0.12	0.56
239Pu	σc	0.05	0.0	- 0.04	- 0.55	- 0.60	0.0	0.02	- 0.40	- 0.87
	σ <sub>in</sub>	0.10	0.11	-	· -	-	0.08	-	-	-
	σ <sub>el</sub>	0.10	0.0	0.01	0,01	0.0	0.0	0.0	0.01	0.0
	ν	0.02	- 0.32		-	-	- 0.20	-	-	-
	σ <sub>f</sub>	0.03	- 0.26	-	-	-	- 0.17	-	-	- '
238 U	σc	0.05	0.07	- 0,29	- 2.12	- 0.78	0.05	0.03	- 1.24	- 0.93
	σ <sub>in</sub>	0.10	1.86	-	-	-	0.78	-	-	-
	σ <sub>el</sub>	0.10	- 0.07	0,15	0.25	0.04	- 0.05	- 0.02	0.11	0.02
	σ <sub>in</sub>	0.10	0.46	-	-	-	0.16	-	-	-
Na	σ <sub>e1</sub>	0.10	0.14	1.65	1.29	0.06	0.05	0.63	0.53	0.04
	σ <sub>c</sub>	0.10	0.01	0.0	- 0.07	- 0.03	0.01	0.01	- 0.11	- 0.10
Fe	σ <sub>in</sub>	0.10	0.36	-	-	-	0.50	_	-	-
· · · · · ·	σ <sub>el</sub>	0.10	0.05	0.28	0.24	0.06	- 0.01	0.27	0.28	0.10
С	σ <sub>e1</sub>	0.10	0.17	1.67	0,95	0.08	-	-	-	-
0	σ <sub>el</sub>	0.10	-	-	-	-	0.16	1.40	0.65	0.14
Be	σel	0.10	-	-	-	-	0.11	0,83	0.40	0.07

TABLE VIII. SENSITIVITY VALUES RELATIVE TO PROMPT NEUTRON LIFETIME (in percent  $\delta \rho_{bil} / \rho_{bil}$ )

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TABLE IX.	SENSITIVITY	VALUES	RELATIVE	то	PROMPT	' NEUTRON	LIFETIME	CONSEQUENT	ON
REAL FLUX	VARIATIONS	(in percen	at $\delta \rho_{\rm bil} / \rho_{\rm bil}$ )					·	

		System		Car	bide			Oxi	ide	
Material	Param. o	Gr. δσ/σ	1	2	3	4	1 .	2	3	4
	ν	0.02	0.0	- 0.01	0.0	0.0	0.0	0.0	0.0	0.0
	σf	0.03	0.0	- 0.28	- 0.33	- 0.16	0.0	- 0.19	- 0.30	- 0.23
239Pu	σc	0.05	0.0	- 0.09	- 0.31	- 0.17	0.0	- 0.06	- 0.28	- 0.25
	$\sigma_{in}$	0.10	0.08	-	-	-	0.06	-	-	· _
	σ <sub>el</sub>	0.10	0.0	0,02	0.02	0.0	0.0	0.01	0.01	0.0
	ע	0.02	0.01	-		-	0.01	-	-	
	σ <sub>f</sub>	0.03	0.04	-	-	-	0.02		° <b>-</b>	-
<sup>238</sup> U	σ <sub>c</sub>	0.05	- 0.01	- 0.71	- 1.32	- 0.24	0.0	- 0.38	- 0.95	- 0.31
	σ <sub>in</sub>	0.10	1.26	-	-	-	0.61	-	-	-
	σ <sub>el</sub>	0.10	0.0	0.33	0.18	0.02	- 0.01	0.15	0.10	0.02
· · · · · · · · · · · · · · · · · · ·	σ <sub>in</sub>	0.10	0,33	-	-	-	0.13	-	-	-
Na	σ <sub>el</sub>	0.10	0,16	1.69	1.02	0.03	0.07	0.69	0.48	0.03
· · · · · · · · · · · · · · · · · · ·	σ <sub>c</sub>	0.10	0.0	- 0.02	- 0.04	- 0.01	0.0	- 0.03	- 0.08	- 0.03
Fe	σ <sub>in</sub>	0.10	0.25	-	-	· -	0.38	-	-	-
	σ <sub>el</sub>	0.10	0.06	0.32	0.17	0.03	0.03	0.37	0.22	0.07
С	σ <sub>e1</sub>	0.10	0.17	1.66	0.74	0,05	-	-	-	-
0	σ <sub>el</sub>	0.10	-	-	-	-	0.20	1.46	0.56	0.11
Ве	σel	0.10	-	-	-	-	0.09	0.82	0.34	0.06

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results of the sensitivity study are shown in Table VIII. The major effects lie, as expected, in the real flux spectrum variations, as indicated in Table IX, where the real flux change effects are presented. For this reason the  $\sigma_{el}$  of light elements and  $\sigma_{in}$  and  $\sigma_c$  of the fertile material are particularly involved. As far as the adjoint flux variations are concerned, the major role is played by the change of  $\nu$ ,  $\sigma_f$  and  $\sigma_c$  of the fissile and fertile materials.

#### 7. CONCLUSION

The generalized perturbation techniques used in this paper seem to represent a powerful tool for analysing integral parameters of interest in the study of fast reactor safety, insofar as they permit a considerable improvement in specific differential data. In this particular analysis, the final values of Tables II, VI and VIII, which summarize the whole work, have not been interpreted in terms of standard deviations of the integral parameters themselves as this would have involved assessing some sort of correlation among different groups, which is beyond the scope of the paper. These results, however, clearly indicate that, among those considered, the integral parameter most sensitive to the assumed uncertainties of cross-sections appears to be the sodium void effect. To reduce its uncertainty as much as possible, a noticeable improvement should be made in our knowledge of the fission and capture parameters of the fissile and fertile materials, and of the elastic and inelastic parameters. An improvement here would have a beneficial effect also on the other integral quantities considered.

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#### DISCUSSION

H. W. KÜSTERS: I am not quite clear about your generalized perturbation method. Your paper states that you have some procedure for accounting for changes in real and adjoint fluxes; now, in trying to improve first-order perturbation it seems better to do two successive  $k_{eff}$ calculations. With your method, can you really save computer time on calculating second-order effects?

M. SALVATORES: Yes, in fact we need do only one  $k_{eff}$  caculation. As I indicated, the solution of the iterative system gives functions  $\psi^*$  and  $\psi$  which enable us to evaluate (by means of Eq.(2) in the paper) the changes in real and adjoint fluxes corresponding to every required parameter variation, without direct calculation of any of these variations. In this way we can save computing time and obtain more information about the effects under investigation.

## A RE-EVALUATION OF <sup>235</sup>U, <sup>238</sup>U AND <sup>239</sup>Pu CROSS-SECTIONS BASED ON MICROSCOPIC AND INTEGRAL DATA

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#### Abstract

A RE-EVALUATION OF <sup>235</sup>U, <sup>238</sup>U AND <sup>239</sup>Pu CROSS-SECTIONS BASED ON MICROSCOPIC AND INTEGRAL DATA. A generalized least-squares method to improve microscopic cross-section evaluations by means of integral data, has been applied to re-evaluate the cross-sections of <sup>235</sup>U, <sup>238</sup>U and <sup>239</sup>Pu, using critical-mass data of 24 simple metallic systems, composed of these isotopes.

It has been found that, after some minor modifications of the original cross-section set, most of the experimental integral data could be reproduced. The cross-section modifications, as a rule, were of the order of a few per cent and well within the uncertainties in the cross-sections. The exception to the rule was the  $^{239}$ Pu fission cross-section in the energy range up to about 150 keV, which had to be decreased by 15-20%. This result independently confirms the recent measurements of White et al.

#### INTRODUCTION

The modification of cross-sections based on results of integral experiments has recently been the subject of several papers [1-5]. These papers, however, only discuss the adjustment of the so-called group constants, these constants having been previously derived in one way or another from a basic set of microscopic cross-sections. But sets of group constants, by their very nature, are usually useful only for well-defined families of multiplying systems and their application is thus rather limited. The notion of <u>universal</u> group constants a priori seems to be self-contradictory. The more fundamental problem in this context should therefore be the adjustment, or rather the improvement of the basic cross-section set itself.

In this note we report the first results obtained by a generalized least-squares procedure to improve microscopic cross-section sets by integral data, proposed by our group [6].

This method essentially reduces to minimizing the form

$$Q = \sum_{i} \int \left( \frac{\sigma - \hat{\sigma}}{\Delta \hat{\sigma}_{1}} \right)^{2} \omega dE + \sum_{i} \left( \frac{\gamma [\sigma] - \hat{\gamma}}{\Delta \hat{\gamma}} \right)^{2}$$

Here the first summation refers to all cross-sections pertinent to the systems considered, and the second to all integral quantities taken into account. The  $\hat{\sigma}$ ,  $\hat{\gamma}$ , etc. are the <u>measured</u> values of the relevant quantities, from which we start, the  $\sigma$  are the modified, unknown cross-sections, and the  $\gamma[\sigma]$  are the <u>calculated</u> values of the integral quantities.

All explicit formulas are derived in Ref.[6], except for a more recent improvement incorporated in the procedure [7] taking account of the total cross-section data, which are generally more reliable than those of partial cross-sections.

#### DESCRIPTION OF INPUT DATA AND RESULTS

As a first practical test of the method, we have applied it to simple spherical critical systems. We had gathered data on critical uranium and plutonium spheres, bare or reflected by natural uranium, and also on bare or reflected systems with an inner plutonium core surrounded by a uranium shell. Thus, except for minor  $^{240}$ Pu and gallium impurities, these systems are comprised of three isotopes only, namely  $^{235}$ U,  $^{238}$ U and  $^{239}$ Pu. We have so far considered 24 such systems, the details of which are collected in Table I.

As our basic microscopic cross-section set, the one which we set out to improve by means of the above critical-mass data, we have chosen the 1962 Karlsruhe set [8]. The relative error of the various partial and total cross-sections have been taken from Schmidt [9].

Plutonium masses quoted include all impurities.  $\delta$ -phase plutonium includes 1 wt  $\frac{1}{\gamma}$  of gallium. Uranium masses are the <sup>235</sup>U masses. All data have been taken from Paxton [16], except those for systems UR 2, 4, 5, 6 and 9 which were taken from Templin [17]. The last two columns give the values of the parameters  $\gamma$ , calculated with the 1962 Karlsruhe cross-section set ( $\hat{\sigma}$ ) and with the modified set ( $\sigma$ ).

	1		Fissionable	Core			Reflec	tor		
System		Pu-239			U-235		Nat. Urs	nium	<b>γ</b> [σ] - 1	Υ[σ] - l
Symbol	Mass (kg)	Density (gr/cm <sup>3</sup> )	Pu-240 (%)	Mass (kg)	Density (gr/cm <sup>3</sup> )	υ238 (%)	Thickness (cm)	Density (gr/cm3)	(x1000)	(x1000)
P	16,28	15.66	4.5						6.783	761
PR1	8.39	15.62	4.9				4.128	18.92	10.488	.180
PR2	6.22	15.58	1.35				11.684	19.00	9.608	•353
# PR3	5.91	15.36	4.8				19.609	19.00		
PR4	5.73	15.63	1.5			1	24.130	19.00		
PU1	8.39	15.62	4.9	12,64	18.80	6.83			4.885	•214
PU2	5.72	15.56	1.5	18,80	18,80	6.82			2.938	•377
#PU3	2.527	19.22	4.7	26,80	18,80	6.83				
#PU4	2.022	15.60	4.7	36.70	18,80	6.83				
<b>#P</b> U5	2.024	15.62	1.5	36.35	18,80	6.83				
PUR1	1.615	19.48	2.34	8.87	18,80	6.8	18.923	19,00	1,326	.446
PUR2	1.610	19.42	4.73	9.09	18.80	6.8	18,872	19.00	1.115	•299
UR1				34.0	18,75	6.1	1.765	19.00	-7.018	378
UR2				30.86	18.80	6.5	2.54	19.00	-4.385	1.378
UR3				24.9	18.75	6.1	4.470	19.00	-3.466	<b>.</b> 898
UR4				24.5	18.75	6.5	4.75	18.80	-3.676	.514
UR5				23.75	18.75	6.5	5.08	18.80	-3.900	.108
UR6				23.56	18.80	6.5	5.08	19.00	-3.546	.469
UR7				19.2	18.75	6.1	8.941	19.00	-3.147	427
URS				18.55	18.75	6.1	9.957	19.00	-3.351	787
UR9				18.42	18.80	6.5	10.16	19.00	-3.440	923
#UR10				16.65	18.62	6.8	18.009	19.00		
Ul				48.8	18.75	6.2			-10.044	913
U2				48.7	18,75	6.1			-10.154	-1.011

TABLE I. DESCRIPTION OF THE 24 CRITICAL SYSTEMS THE DATA OF WHICH WAS USED TO IMPROVE THE MICROSCOPIC CROSS-SECTION EVALUATION

#### SM-101/67

The integral quantities that we consider, the  $\gamma$ 's of our problem, are obviously the reactivities, or the multiplication constants, of the above-mentioned systems. We find it more convenient, however, to employ other, closely related quantities, namely the eigenvalues of the equation

$$\vec{\Omega} \cdot \nabla \Psi + \Sigma \Psi = \frac{1}{\gamma} S$$

The  $\gamma[\sigma]$  of the critical systems have been calculated in a 25-group  $S_8$  approximation. The experimental values, by definition, are all  $\hat{\gamma} = 1$ . As for the errors  $\Delta \hat{\gamma}$ , these are often (but not always) given along with the other experimental results, but the values quoted vary rather suspiciously from system to system. We have therefore, somewhat arbitrarily, assumed that  $\Delta \hat{\gamma} = 0.001$  for all the systems. This is equivalent to 50 - 100 g plutonium in the Pu-core systems, and to 250 - 500 g uranium in the U-core systems.

#### TABLE II. CORRECTION FACTORS FOR <sup>235</sup>U.

The second column gives upper bounds of energy intervals over which the factors are constant. In the original set the relation  $\overline{\nu}(E) = 2.25 \pm 0.132 E$  was used.

1				p (x100)		
No.	Energy MeV	Pission	Capture	Inelastic Scattering	Elastic Scattering	$\overline{\nu}$
			•			
1	•020	•5	-1,2	•0	•1	•0
2	•040	2.4	-2.5	•0	1.1	•5
3.	<b>,</b> 070	3.3	-2.6	•1	1.9	•6
4	.090	3.3	-2,1	.4	2.6	•6
5	.120	- 3•4	-1.8	.8	2.9	.5
6	.160	3.5	-1.5	1.3	3.0	•5
7	.200	3.7	-1.3	1.9	2.9	•5
8	<b>.</b> 250	3.9	-4.8	2.5	2.7	•6
9	.300	4.0	-4.1	3.1	2.2	.6
10	•375	4.1	-3.5	3.7	1.6	•5
11	.450	4.0	-3.0	4.0	1.1	.5
12	.550	3.9	-2.7	3:9	.7	.5
13	<b>.</b> 650	4.3	-2.5	3.7	.3	.5
14	.750	4.0	-2,2	3.4	.1	•4
15 <sup>°</sup>	.900	3.7	-1.7	2.9	1	•4
16	1.100	3.4	-1.3	2.1	2	•4
17	1.300	3.0	9	1.5	3	.4
18	1.450	2,8	7	1.2	3	.3
19	1.550	2,6	6	1,2	3	•3
20	1.650	2,5	5	1.2	3	.3
21	2.0	2,2	4	1.1	4	•2
22	2.4	1.8	3	.9	3	•2
23	3.0	•6	1	.9	2	•3
24	4.0	•4	1	•6	1	•2
25	14.0	•0	0	•1	0	•0

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#### TABLE III. CORRECTION FACTORS FOR <sup>238</sup>U.

The second column gives upper bounds of energy intervals over which the factors are constant. In the original set the relation  $\overline{\nu}(E) = 2.48 \pm 0.132 E$  was used.

NO.	Snergy	p (x100)						
	MeV	Fission Capture		Inelastic Scattering	Elastic Scattering	$\overline{\nu}$		
1	.02	•0	•5	.0	6	.0		
2	.04	.0	.3	.0	-1.4	.0		
3	.07	.0	1.1	1	-2.5	.0		
4	.09	.0	3.4	6	-3.4	.0		
5	•12	••	4.5	-1.8	-6.8	.0		
6	.16	.0.	5.2	-3.6	-9.9	•		
7	.20	.0	5.3	-2.9	-10.0	.0		
8	.25	• .0	.8	-3.8	-9.4	.0		
9	.30	٥.	.7	-4.6	-8.5	.0		
10	.375	.0	.6	-5.3	-7.3	.0		
11	.450	.0	•5	-5.7	-6,1	.0		
12	.550	.0	•5	-5.9	-5.0	.0		
13	.650	.0	.5	-7.5	-3.9	0		
14	.750	0	.4	-6,2	-2.5	0		
15	.90	0	.4	-4.9	-1.1	0		
16	1.1	1	.4	-4.3	7	0		
17	1.3	-,1	.2	-2,9	6	~.0		
18	1.45	3	.1	-2,5	5	~.0		
19	1.55	6	.1	-2.0	5	~.1		
20	1.65	7	1	-2.6	-,2	1		
21	2.0	8	.1	-2.3	1	1		
22 .	2.4	-1.9	.0	-1.3	.1	~.1		
23	3.	-1.4	.0	-1.2	.2	0		
24	4.	9	.0	9	.2	~.0		
25	14.	0	.0	1	.0	0		

The neutron angular fluxes and adjoint angular fluxes have also been calculated in the above-mentioned approximation and have then been used as input to a multigroup perturbation code to obtain the derivatives  $\partial \gamma / \partial \sigma_g$  which are necessary for our analysis.

Finally, by the explicit formulas of Ref.[6] (slightly modified as already mentioned), the relative correction factors defined by

obtained for each partial cross-section and for  $\overline{\nu}$ . The p's are given in Tables II, III and IV. The original and modified microscopic crosssections are given in Figs 1, 2 and 3.

As will be explained below, only 18 systems were used to obtain the modified cross-section set. The values of the 18 corresponding  $\gamma$ 's, calculated with the original (1962 Karlsruhe) and modified sets of cross-sections, are also given in Table I.

	Endrgy MeV	p (x100)						
No.		Fission	Capture	Inelastic Scattering	Elastic Scattering	$\overline{\nu}$		
1	.02	-5.7	10.9	•2	•5	6		
2	.04	-15.9	22.2	.6	2,4	-1.6		
3	.07	-22.5	16.9	1.2	4.5	-2.3		
4	.09	-23.6	14.8	1.1	5.7	-2.4		
5	.12	-14.6	9.5	.4	4.7	-2.3		
6	.16	-3.5	3.0	4	3.3	-2.1		
7	.20	-3.1	2.6	5	3.2	-1.9		
8	.25	-2,8	1.9	7	3.0	-1.6		
9	.30	-2.4	1.4	-1.0	2.8	-1.4		
10	.375	-2.0	1.1	-1.2	2.6	-1.2		
11	.45	-1.7	.8	9	2.3	-1.0		
12	.55	-1.4	.6	5	2,0	8		
13	.65	-1.2	.4	0	1.8	7		
14	.75	-1.0	.3	.0	1.7	6		
15	.90	9	.2	.1	1.5	5		
16	1.10	7	.1	.3	1.3	4		
17	1,30	-1.2	.2	.6	1.3	3		
18	1.45	-1.0	.1	.7	1.1	3		
19	1.55	8	.1	.9	.9	3		
20	1.65	7	.1	1.0	.8	2		
21	2.0	5	.1	1.1	.6	2		
22	2.4	4	.0	1.0	.4	1		
23	3.0	3	.0	.8	.2	1		
24	4.0	2	.0	.6	.0	1		
25	14.0	0	.0	.0	.0	0		

TABLE IV. CORRECTION FACTORS FOR <sup>239</sup>Pu.

The second column gives upper bounds of energy intervals over which the factors are constant. In the original set the relation  $\overline{\nu}(E) = 2,89 \pm 0,123 E$  was used.

#### DISCUSSION

It should be clear from the tables and figures that the cross-section modifications are generally very small indeed. This actually implies that the experts who analysed the multitude of direct cross-section measurements and theoretical information and derived the 'best' crosssection set have done a pretty good job. With some more luck they could just as well have derived most of the modified set, which seems to be almost as consistent with the primary data as the original set is. Since the integral quantities are often extremely sensitive to variations in the cross-sections, rather slight changes in the cross-section curves might greatly improve the agreement between the calculated and experimental values of the  $\gamma$ 's. In other words, the experimental cross-section and integral data are, in fact, consistent with each other. Yet, because of the enormous scatter of the former and the extreme sensitivity of the latter, it is practically impossible at present to derive satisfactory cross-section curves from the cross-section data alone. The direct



FIG. 1. Microscopic cross-sections of  $^{235}$ U. — 1962 Karlsruhe set ( $\partial$ ); - - - modified cross-sections ( $\sigma$ ) resulting from the least-squares analysis including critical-mass data of 18 systems.

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FIG.2. Microscopic cross-sections of  $^{238}$ U. — 1962 Karlsruhe set (ô); --- modified crosssections ( $\sigma$ ) resulting from the least-squares analysis including critical-mass data of 18 systems. SM-101/67



FIG. 3. Microscopic cross-sections of  $^{239}$ Pu. —— 1962 Karlsruhe set ( $\hat{\sigma}$ ); - - - modified crosssections ( $\sigma$ ) resulting from the least-squares analysis including critical-mass data of 18 systems;  $- \Phi$  measurements of White et al. [12].

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cross-section measurements do supply an excellent 'spine' for the curves, but an additional guide is essential for obtaining their finer structure. The integral measurements, by their superior accuracy, supply this guide for drawing the 'ripples' superposed on the 'spine'.

In some cases, however, the cross-section modifications are neither small nor lie within the experimental uncertainties in the original curves. The case in point in the present analysis is the fission crosssection of <sup>239</sup>Pu. This exception, however, is quite remarkable. It is now generally accepted [10, 11] that the earlier evaluations of this cross-section up to about 150 keV should be substantially modified in view of the new measurements of White et al [12] given in Fig. 3. This is exactly what is indicated by our results. Had our results been obtained before the publication of these measurements, we would have suggested that the original fission cross-section in this energy range should be rechecked. The fact that the only real deviation of a modification crosssection from the original curve had already been discovered by direct measurements seems to fully vindicate the re-evaluation procedure. In the few other cases where the modifications are not really small, amounting to about 10% or more, the uncertainties in the original cross-sections are rather large, and the modifications always lie within limits.

In the final analysis, the modified cross-section set is based on 18 systems only. Their  $\gamma$ 's, calculated with the modified set, nicely agree with their experimental values: they are all equal to unity within 1% or less. It turned out that, as soon as part or all of the other six systems, marked by an asterisk in Table I, were included, many of the resulting correction factors started to 'jump' and more often than not reached values far exceeding the input uncertainties. At the same time, while the nice fit of the first 18  $\gamma$ 's was not appreciably affected, the  $\gamma$ 's of the additional systems persistently refused to fall into line. This seems to indicate that, while the 18 measurements are consistent with the direct cross-section data and with each other, the other six are definitely neither consistent with the 'differential' nor with the rest of the integral data.

We have also applied the least-square procedure to different combinations of parts of the 18 consistent systems. The cross-section sets obtained in each case, even when only as few as three systems (P, PR1 and UR4) were considered, practically coincided with the set obtained by using all 18 systems. It is therefore evident that additional criticalmass data on fast plutonium and uranium systems of the kind considered in this analysis will be adequately reproduced by the present modified cross-section set. Moreover, the inclusion of additional critical-mass data of such systems will not contribute much to further improvement of the cross-sections.

An interesting coincidence is worth mentioning. All the systems excluded from the final analysis, except PR4 (POPSY), are quoted as 'private communications' only, which fact might imply that the measurements quoted were not final. As for POPSY, this is of course not really a spherical system and our analysis may indicate that the quoted dimension of its equivalent spherical reflector is in error.

We would also like to remark that in some cases we could not use published data [13-15] for lack of information as to the isotopic composition of the systems.

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#### DISCUSSION

B.I. SPINRAD: I notice that you started with two quite different cross-section sets, the Parker and Schmidt sets, and that your results indicate corrections to the Schmidt set. When you do the same to the Parker set, do the corrected results show any convergence?

Y. YEIVIN: As I stressed in my talk, the modifications of the cross-section curves are usually in the form of 'ripples', superposed on a basically correct 'spine'. Since the two cross-section sets sometimes differ quite markedly we cannot expect the respective modified sets to coincide. However, Fig.A shows what we may achieve by modifying the cross-sections: the solid line joins the points denoting the reactivities of the 18 systems, calculated with Schmidt's cross-

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section set; and the dashed line refers to Parker's<sup>1</sup> set. Once this procedure has been applied to each set, the reactivities calculated with the modified sets agree with the experimental values to within  $\pm 2.5\%$ ; it becomes clear that the German plutonium cross-sections are 'over-reactive', while the British ones are 'under-reactive'. You will recall



FIG.A. Comparison of calculations using Schmidt's cross-section set with Parker's 1960 set.

that the German fission cross-section was decreased appreciably (in agreement with the measurements of White et al.), thereby bringing down the reactivities of the plutonium systems. One might expect furthermore that the British fission cross-section would increase, where-

<sup>&</sup>lt;sup>1</sup> BUCKINGHAM, B.R.S. et al., UKAEA Rep. AWRE 0-28/60 (1960).

as in fact it also decreased when the modification procedure was applied to the British set. It seems, therefore, that whereas significantly different cross-section sets will not really converge upon modification by our procedure, the indicated trends will still apply.

J.Y. BARRE: How many multiplying systems containing <sup>239</sup>Pu did you use in your modification? How many of these systems have an energy spectrum sufficiently degraded to permit correction of the <sup>239</sup>Pu fission cross-section in the 10-keV region?

Y. YEIVIN: Of the 18 systems used to modify the cross-sections (details of which are given in Table I of paper SM-101/21), one was a bare Pu sphere (JEZEBEL), two were Pu spheres reflected by natural uranium, two were bare systems with an inner Pu core surrounded by a  $^{235}$ U shell, and two were mixed systems with a natural uranium reflector. The other 11 systems had a uranium core. Obviously, all of these are hard-spectrum systems, not very sensitive to the cross-section values at 10 keV or below. You are right in implying that softer-spectrum systems should be included in the analysis, in order to improve the cross-sections in the low-energy region, and it is our intention to include such systems in future analyses.

E.D. PENDLEBURY: What standard deviations did you assume for the different  $k_{eff}$  values?

Y. YEIVIN: The errors in the critical masses are not always quoted in the literature, and even when they are we find that the quoted values vary over a wide range for the various systems. We therefore assumed, somewhat arbitrarily, that the uncertainties of the measured reactivities were 2.5% throughout, which is equivalent to 50 - 100 g plutonium, or to 250 - 500 g uranium for the respective systems. These limits are also marked on Fig.A.

# EXPERIMENTAL METHODS

(Session III, Part 2)

Chairman: K.E.J. WIRTZ

### A MODIFIED LEAST-SQUARES METHOD TO UNSCRAMBLE PROTON RECOIL SPECTRA

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#### Abstract

A MODIFIED LEAST-SQUARES METHOD TO UNSCRAMBLE PROTON RECOIL SPECTRA. A computer program to UNFOLD proton recoil spectra has been written for the IBM/360-30 computer. By a least-squares technique it determines the neutron spectrum as weights in the sum of theoretically derived response functions when fitted to the measured recoil spectrum. Physically unreasonable oscillations, which easily result from this type of analysis, are depressed by adding a term  $y S_2$  to the usual sum  $(S_1)$  of squared deviations before minimizing. Here  $S_2$  is a measure of how well the points in the neutron spectrum fit polynomials of the k:th degree drawn through k+2 points. k equals 2 or 3 has been used. The relative weights of the two sums are adjusted by means of y.

The program has been tested on pulse-height distributions obtained by calculations from a typical fast reactor neutron spectrum. The response functions used apply to a spherical proportional counter and take into account wall effects and counter resolution. It turns out to be possible to reproduce a neutron spectrum with the high cut-off neutron energy corresponding to proton ranges of the order of the counter radius. The program is also capable of correcting for proton pulses due to neutrons above the cut-off energy, provided that the shape of the neutron energy spectrum is known for these neutrons. In regions where the spectral variations are rapid compared to the counter resolution a smoothed result is obtained.

It is found that the best reproduction of the original neutron spectrum is obtained for the  $\gamma$ -value which gives  $S_1$  equal to the number of points in the recoil spectrum. An estimate of the statistical errors in the neutron spectrum thus obtained is given. A typical computing time for one  $\gamma$ -value is about 2.5 min when 53 neutron energy and 71 proton energy points are used.

#### 1. Introduction

Hydrogen-filled proportional counters have proved to be suitable for measuring neutron spectra in fast reactors in the energy region between a few keV and a few MeV [1, 2]. These detectors, however, show a deviation from the ideal rectangular response function due to wall effects in the high energy region and to the limited energy resolution (about 10 %).

Several methods have been used for unscrambling measured pulse-height distributions once the response function of the experimental apparatus is known. In the case when the response has the ideal rectangular shape, the neutron spectrum is easily obtained by differentiation. The most direct way to take the actual shape of the response function into account is perhaps to make an inversion of the response matrix, but the solution thus obtained often exhibits physically unrealistic oscillations. Brunfelter et al. [3] have shown how these can be reduced by using a special form of the matrix. Burrus et al. [4] have described a way of overcoming the oscillations by accepting only non-negative solutions and Yates [5] has given a method including minimization of a function, depending both on the goodness of fit and on the smoothness of the solution. A similar method has been suggested by Grissom et al. [6] under the assumption of Gaussian response. Our investigation shows how the latter method can be extended and applied to pulse-height distributions from proton recoils in a spherical proportional counter when both wall effects and resolution are taken into account.

#### 2. Presentation of the method

When the spectrometer is irradiated in a neutron spectrum  $\phi(E_n)$  the corresponding pulse-height distribution,  $P(E_p)dE_p$ , defined as the number of pulses with pulse-heights between  $E_p$  and  $E_p + dE_p$ , will be

$$P(E_p)dE_p = \int_{0}^{E_{nmax}} \phi(E_n)\Sigma(E_n) G(E_n,E_p)dE_pdE_n$$
(1)

where

$$E_{n} = neutron energy$$

$$E_{nmax} = maximum neutron energy in the actual neutron field$$

$$\Sigma(E_{n}) = total efficiency of the spectrometer at energy E_{n}$$

$$G(E_{n},E_{p})dE_{p} = probability that a neutron of energy E_{n} will give a pulse in the spectrometer with pulse-height between E_{p} = dE_{p} + dE_{p}$$

The integral in (1) is approximated with a Simpson's rule numerical quadrature. In order to get appropriate spacing in the summation, the energy variable is expressed in lethargy units. Thus Eq. (1) will be

$$P_{j} = \sum_{i=1}^{n} \phi_{i} \Sigma_{i} G_{ij} C_{i} \Delta_{i}$$
(2)

where

C; = weights used in the numerical quadrature

 $\Delta_i$  = step-lengths in lethargy units in the numerical quadrature

and the rest of the abbreviations are obvious.

If we now want to fit a measured pulse-height spectrum  $\hat{P}_j(1 \le j \le m)$  in the least squares sense to Eq. (2), this is equivalent to minimizing the sum

$$s_{1} = \sum_{j=1}^{m} \frac{1}{(\Delta \hat{P}_{j})^{2}} (\hat{P}_{j} - \sum_{i=1}^{n} \phi_{i} \Sigma_{i} G_{ij} C_{i} \Delta_{i})^{2}$$
(3)

where  $\Delta \hat{P}_{i}$  is the statistical uncertainty in  $\hat{P}_{i}$ .

The solution  $\{\phi_i\}$  thus obtained will usually exhibit physically unrealistic oscillations, due to the badly conditioned matrix.

In order to damp these oscillations, a term  $\gamma \cdot S_2$  is in the present method added to  $S_1$  before minimizing.  $S_2$  is here defined as n-1  $\sum_{i=k+1}^{n-1} R_{ki}^2$  where  $R_{ki}$  is the rest term in an approximate Taylor series exi=k+1 is constant of the solution  $\phi_{i+1}$  in the k previous  $\phi_i$ :s (see Ref. [6]). If k = 2, for example, this means that one is looking for a solution where the  $\phi_i$ :s four by four closely follow parabolas when they at the same time satisfy Eq.(1).  $\gamma$  is a parameter that adjusts the relative weights put on  $S_1$  and  $S_2$ , respectively.

The set  $\{\phi_i\}$  will be obtained by the n linear equations

$$\frac{\partial (S_1 + \gamma S_2)}{\partial \phi_i} = 0 \qquad i = 1 \dots n \tag{4}$$

The sum  $S_1$  ought to be of the order of the number of measured points (m) when a physically realistic solution to the problem is found since the terms are weighted with their statistical uncertainties. A search is therefore made in the program by varying the parameter  $\gamma$  until  $S_1 = m$ .

The statistical uncertainties in the solution are obtained from the formula

$$(\Delta \phi_{i})^{2} = \sum_{j=1}^{m} \left( \frac{\partial \phi_{i}}{\partial P_{j}} \right)^{2} (\Delta \hat{P}_{j})^{2}$$
(5)

These uncertainties are easily obtained from the same matrix that gives the solution to Eq. (4).

#### 3. Response functions

**^** .

Snidow [7] has shown that it is possible to calculate a response of spherical counters which takes wall effects into account and which well reproduces measured response functions. We have used his method on a spherical counter 3.94 cm in diameter, filled with 7 atm CH<sub>4</sub>, and determined the response functions  $G_1(E_n, E_p)$  for neutron energies in the region between 66 keV and 10 MeV. The finite resolution of the counters can be taken into account by assuming Gaussian resolution function and defining the final response function as

$$G(E_{n},E_{p}) = \int_{E_{0}}^{E_{n}} dE \cdot \frac{1}{\sigma(E)\sqrt{2\pi}} e^{-\frac{(E-E_{p})^{2}}{2(\sigma(E))^{2}}} G_{1}(E_{n},E)$$
(6)

(The low integration limit  $E_0$  has to be > 0 in order to avoid singularities.)

In order to speed up the numerical calculations and facilitate interpolations, two approximations to Eq. (6) have been made. The first is to approximate  $G_1(E_n,E_p)$  with a parabola, the second to take the resolution effects into account only close to the step in  $G_1(E_n,E_p)$  at  $E_p = E_n$ . The following modified form of Eq. (6) has been used:



The coefficients A, B and C are determined by a least squares fit to the response functions calculated according to Snidow with the further restriction that the total area should remain unchanged. The resolution function  $\sigma(E)$  is for the time being assumed to be proportional to the energy, but it is easy to include an energy dependence, caused for instance by electronic noise. Fig. 1 shows  $G(E_n, E_p)$  according to Eq. (6) and the difference between the results obtained with Eq. (6) and Eq. (6a) for the neutron energies 0.63 and 3 MeV. The latter is the highest energy used in the unscrambling procedure, and here the approximations made are largest.

#### 4. Test spectrum

In lack of an experimental pulse-height spectrum and in order to make it possible to control the different factors affecting the result of the unfolding procedure a test pulse-height spectrum has been constructed using Eq. (1). A theoretical neutron spectrum (Fig. 2) calculated for core 7 of the FRO reactor (cf. Table I) with the SPENG program [8] was used as input together with response functions calculated with  $\sigma = 0.0423$  E according to Sect. 2.2, which corresponds to FWHM of 10 % for a Gaussian peak. Since it is essential that no numerical uncertainties introduced when evaluating Eq. (1) will affect the conclusions drawn from the calculations, the number of neutron energy and pulse-height points has been much larger during the construction of the test spectrum than in the unscrambling procedure. Statistical uncertainties to the pulse-height spectrum have been added with the aid of a set of normally distributed random numbers.

#### 5. Results and discussion

The result of an application of the unscrambling program (UNFOLD) [9] to the test spectrum is shown in Fig. 2. The lethargy step in the pulse-height spectrum was taken as 0.03 ( $\frac{E_j}{E_{j-1}}$  = 1.03) and in the final neutron spectrum 0.04 ( $\frac{E_i}{E_{i-1}}$  = 1.04). This corresponds to 129 Eqs. of type (2) with 97 unknowns  $\phi_i$  in the energy region investigated (66 keV to 3 MeV). Since these



FIG.1. Computed response functions  $G(E_n, E_p)$  of a spherical proportional counter (diameter 3.94 cm) filled with 7 atm CH<sub>4</sub>. In the upper figure  $E_n = 0.63$  MeV, in the lower  $E_n = 3$  MeV. The dashed-dotted curves give the results according to Snidow [7], the full line the results according to Eq. (6) and the dashed curve the difference between the results according to Eq. (6a) and Eq. (6). The parameters used in Eq. (6a) were in the two cases:

A = 1.068, B = -0.217 MeV<sup>-1</sup> and  $|C| < 2 \times 10^{-3} \text{ MeV}^{-2}$ A = 2.088, B = -0.88 MeV<sup>-1</sup> and C = 7.74 × 10<sup>-2</sup> MeV<sup>-2</sup>.

numbers exceed the memory capacity of the computer used (IBM 360/30), the unscrambling was divided into two parts. Moreover, pulses due to neutrons above 3 MeV have to be taken into account. At these energies the wall effects are so large that the counter cannot be used to obtain detailed spectral information. Instead the shape of the neutron spectrum at these energies is assumed to be known and attached to the solution below 3 MeV. The pulses due to the neutrons are included when a fit is made in the region between 400 keV and 3 MeV as described in Sect. 2. The pulse-height spectrum below 500 keV due to neutrons above this energy is then calculated from Eq.(1) and subtracted from the original pulse-height spectrum and an unscrambling in the low energy region is started. Uncertainties in the result are introduced at both ends of the energy region investigated since the information here is incomplete due to the finite resolution. For that reason the two parts of the unfolding are made to overlap.



FIG.2. Comparison between input neutron spectrum (full line) and resulting neutron spectrum (crosses) obtained with the UNFOLD program. The points above 450 keV were obtained directly, the points below this energy after subtracting of pulses due to higher energy neutrons as discussed in the text.

TABLE I:	Atomic	Densities	in	FRO	Core	7

Isotope	U <sup>235</sup>	U <sup>238</sup>	Ni	Cr	Fe	н	G	F
$10^{22} \text{ at/cm}^3$	0.354	1.395	2.044	0,096	0,406	0.599	2,150	0,006



FIG. 3. Comparison between input neutron spectrum (full line) and resulting neutron spectrum obtained by differentiation. The crosses apply to the case when pulses due to neutrons above 3 MeV were included, the points to the case where they were excluded.

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UNFOLD has been applied to the same pulse-height spectrum both with and without statistical uncertainties added. The results shown in Fig. 2 correspond to a total number of counts of about  $3 \cdot 10^6$ . In the present application the pulse-height spectrum was regarded as a point-spectrum, but a run in which each point represented an integration over a narrow interval did not change the results appreciably.

The result of a direct differentiation of the same pulse-height spectrum is shown for comparison in Fig. 3. Since only two adjacent pulse-height points are at present used to obtain each point in the neutron flux the scattering due to statistical uncertainties will be large. Because of this we only show the case where no uncertainties have been added. We show both the case with and without pulses due to neutrons above 3 MeV added. As is to be expected the differentiation fail to reproduce the high energy part of the neutron spectrum where the wall effects in the counter are appreciable. Below about 600 keV (A = 1.06 in the Eq. (6a)) the obtained spectrum follows the input spectrum better but is still somewhat high and fine details are smeared out.

The result of UNFOLD given in Fig. 2 shows quite satisfactory agreement in the high energy region indicating that it would be possible to extend the usable range of the counter to even higher energies. In the low energy region, however, the spectral details are completely smeared out. When no statistical uncertainties were included a result similar to that obtained by differentiation was obtained. In this case the y-value that gave a minimum in  $\gamma$  S2/S1 was used since this criterion empirically has been shown to give approximately the same y as the statistical one. If a lower value of  $\gamma$  than that giving S1 = m was used when uncertainties were added, oscillations in the obtained spectrum with a period of about 6 energy points start to appear. The results shown apply to k = 2 in the Taylor series expansion, but similar effects were obtained for k = 3. Attempts were made to get better spectral resolution by using more energy points but still without success. The failure of UNFOLD to reproduce the spectral details of the test spectrum is probably due to the fact that peaks and valleys follow each other within one resolution width of the counter. In the errors shown in Fig. 2 and calculated according to Eq. (5) the smoothing effect of the method is included explaining the small errors obtained in the low energy region.

#### 6. Conclusions

In the preceding section it is shown that the proposed method to unscramble pulse-height spectra obtained with spherical proportional counters is capable of extending the useful range of these counters to high energy regions where the wall effects are appreciable. The smoothing condition used to damp unwanted oscillations in the result is too rigorous to allow wanted details in the neutron spectrum to be obtained. Further work to loosen this condition and thus find ways to include known information about the counter resolution is planned. For the time being the best combination seems to be the use of the UNFOLD program in the high energy region and a differentiation scheme in the low energy part.

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#### DISCUSSION

D. STEGEMANN: Have you compared your unscrambling technique with the techniques applied to date using Monte Carlo techniques, so as to correct for wall and end effects?

J. KOCKUM: No, apart from calculations of the response-functions themselves, which have been compared by Snidow and found to give very good agreement.

D. STEGEMANN: What is your assessment of the accuracy obtainable between 1 and 3 MeV with your unscrambling method for smooth and resonance-containing neutron spectra?

J. KOCKUM: The accuracy is very good for a smooth spectrum, as shown in Fig.3. Well separated resonances can probably be resolved, but if two resonances lie within the resolution of the counter they will be smoothed out.

J.W. WEALE: I should like to draw the attention of participants to a report dealing with unscrambling techniques, reference No. AWRE 07/67, which will be available from Aldermaston in about one month's time.
# MESURE EN PILE DE REPONSES EN ENERGIE DE TRITONS OBTENUS PAR LA REACTION <sup>6</sup>Li(n, t) <sup>4</sup>He ET DETECTES AU MOYEN DE DETECTEURS A SEMI-CONDUCTEURS\*

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# Abstract — Résumé

IN-PILE MEASUREMENTS OF THE ENERGY RESPONSES OF TRITONS OBTAINED BY THE <sup>6</sup>Li(n, t)<sup>4</sup>He REACTION AND DETECTED WITH SEMI-CONDUCTING DETECTORS. Neutron spectrum measurements by the <sup>6</sup>Li(n, t)<sup>4</sup>He reaction, using either the parameter  $E_{t}$  or the parameter  $E_{\alpha}$  +  $E_{t}$ , call for knowledge of the distributions – characteristic for the spectrometer and the measurement medium – of the triton energies for monoenergetic neutrons.

These distributions usually have to be calculated from the differential angular cross-sections for triton emission at the neutron energies considered, when the angular distribution of the incidental flux is known. By determining these distributions experimentally in an isotropic neutron flux, one can check or complete the data for the differential angular cross-sections for triton emission.

The paper also shows that by making measurements with collimated detection geometry and comparing the results with those applicable to isotropic conditions, it is similarly possible, in the general case, to obtain an indication of the angular neutron-flux distribution, and to use this information for more precise interpretation of the spectra deduced from the parameters  $E_{\alpha} + E_t$  and  $E_t$ .

The energy distributions of tritons for neutrons with energies of about 250 keV was measured in the leakage spectrum of the HARMONIE reactor. This was done by using a single channel analyser to select a narrow-energy band in the  $E_{\alpha}$  +  $E_{t}$  distribution measured, and measuring the energy distribution of tritons coinciding with pulses from the single channel analyser.

The resolution of the neutron energy selected depends on the half-height width of the  $E_{\alpha} + E_t$  response for thermal neutrons (90 keV for  $30 \mu g$  of lithium fluoride) and the width of the single channel (40 keV).

MESURE EN PILE DE REPONSES EN ENERGIE DE TRITONS OBTENUS PAR LA REACTION <sup>6</sup>Li(n, t)<sup>4</sup>He ET DETECTES AU MOYEN DE DETECTEURS A SEMI-CONDUCTEURS. La mesure de spectres de neutrons au moyen de la réaction <sup>6</sup>Li(n, t)<sup>4</sup>He, soit par le paramètre  $E_t$ , soit par le paramètre  $E_{\alpha}$  +  $E_t$ , nécessite la connaissance des distributions, caractéristiques pour le spectromètre et le milieu de mesure, des énergies des tritons pour des neutrons monoénergétiques. Ces distributions doivent normalement être calculées sur la base des sections efficaces angulaires différentielles d'émission des tritons aux énergies neutroniques considérées, lorsque la distribution angulaire du flux incident est connue. La détermination expérimentale de ces distributions dans un flux neutronique isotrope permet de vérifier ou de compléter les données relatives aux sections efficaces angulaires différentielles d'émission des tritons. On montre également que, par mesure en géométrie de détection collimatée et comparaison au cas isotrope, on peut de même obtenir dans le cas général des indications sur la distribution angulaire du flux de neutrons et les utiliser ensuite pour une interprétation plus précise des spectres déduits à partie des paramètres  $E_{\alpha} + E_t$  et  $E_t$ . On a mesuré la distribution des énergies de tritons pour des neutrons d'énergie voisine de 250 keV, dans le spectre de fuite du réacteur HARMONIE. Pour ce faire, on sélectionne à l'aide d'un monocanal, dans la distribution  $E_{\alpha}$  +  $E_{t}$  mesurée, une bande étroite en énergie et on mesure la distribution des énergies de tritons en coincidence avec les impulsions provenant du monocanal. La définition sur l'énergie des neutrons choisie est déterminée à partir de la largeur à mi-hauteur de la réponse  $E_{\alpha}$  +  $E_{t}$  obtenue aux neutrons thermiques (90 keV pour 30µg de fluorure de lithium) et de la largeur du monocanal (40 keV).

<sup>\*</sup> Travail effectué au Centre d'études nucléaires de Cadarache (CEA), dans le cadre du contrat Euratom «Projet Neutrons Rapides»

# 1. INTRODUCTION

La détermination d'un spectre neutronique à l'aide de la réaction <sup>6</sup>Li(n, t) <sup>4</sup>He peut être fondée sur la mesure, à l'aide de semi-conducteurs, de la distribution des particules alpha et des tritons émis en coincidence (paramètre  $E_{\alpha} + E_t$ ) ou sur la mesure de la distribution en énergie des tritons émis (paramètre  $E_t$ ), principalement pour des énergies neutroniques inférieures à 500 keV [1].

L'interprétation de la réponse en fonction de  $E_{\alpha} + E_t$  nécessite la connaissance de la section efficace totale de la réaction et, comme élément de correction, de la section efficace différentielle d'émission des tritons pour déterminer le rendement du spectromètre en fonction de l'énergie.

Pour la réponse en fonction de  $E_t$ , la section efficace angulaire différentielle constitue l'élément essentiel pour calculer les distributions en énergie des tritons en réponse à des neutrons monoénergétiques.

Le rendement et les distributions en énergie des tritons sont caractéristiques du spectromètre et du milieu de mesure; si, lors de leur détection, les particules alpha et les tritons émis font l'objet d'une collimation, ce rendement et cette distribution dépendent essentiellement de la distribution angulaire des neutrons.

La distribution angulaire étant généralement mal connue nous proposons une méthode de mesure permettant de déterminer expérimentalement, pour un spectromètre et un milieu de mesure donnés, les distributions en énergie des tritons pour des neutrons monoénergétiques, d'où peut être déduit le rendement dans le cas de la méthode  $E_{\alpha} + E_{t}$ .

Dans l'hypothèse où la distribution angulaire des neutrons ne varie pas en fonction de l'énergie des neutrons, il suffit de mesurer la distribution en énergie des tritons correspondant à une seule énergie neutronique pour pouvoir calculer les autres distributions.

Pour une collimation parfaite des particules alpha et des tritons on peut déduire, par la même méthode, la distribution angulaire des neutrons en fonction de l'angle entre le neutron et l'axe de collimation des particules alpha et des tritons.

# 2. PRINCIPE DE LA METHODE

Le spectromètre consiste en un sandwich de détecteurs à semi-conducteurs avec un dépôt de <sup>6</sup>LiF monté entre les deux. Suivant le but recherché la détection des particules alpha et des tritons est collimatée ou non.

La technique combine la mesure du spectre  $E_t$  et  $E_{\alpha} + E_t$ . Nous sélectionnons à l'aide d'un monocanal dans le spectre  $E_{\alpha} + E_t$  une bande étroite en énergie et mesurons la distribution  $E_t$  en coïncidence avec les impulsions provenant du monocanal.

Le schéma électronique de base est donné dans la figure 1. Il peut être complété avec un circuit de coincidence rapide dans la chaîne de sommation pour réduire le bruit de fond  $\gamma$ . La connection 2 du schéma permet de mesurer la bande d'énergie effectivement sélectionnée par le monocanal.

Le degré de collimation, l'épaisseur du dépôt de <sup>6</sup>LiF et la largeur en keV du monocanal sont fixés en tenant compte de la valeur du flux neutronique instantané et du temps de comptage tout en essayant de conserver à l'ensemble des performances optimales.



FIG.1. Schéma électronique.



FIG.2. Dispositif expérimental.

TABLEAU I. LARGEURS A MI-HAUTEUR, EXPERIMENTALES ET THEORIQUES, DES DISTRIBUTIONS  $E_{\alpha} + E_t$ ,  $E_t$  ET  $E_{\alpha}$  POUR UN DEPOT DE <sup>6</sup>LIF D'EPAISSEUR NOMINALE EGALE A  $30\mu g/cm^2$ 

	Eα (keV)		E <sub>t</sub> (keV)		$E_{\alpha} + E_{t}$ (keV)	
	Exp.	Théor.	Exp.	Théor.	Exp.	Théor.
Détection 2 π Détection collimatée	135 95	57 49	55 40	41 40	95	61

#### 3. MESURES

#### 3.1. Montage

Nous avons mesuré le spectre de fuite contre le cœur d'HARMONIE en position haute et fonctionnant à 35 W. Le spectromètre était monté de telle façon que l'axe de collimation était perpendiculaire à la face de la pile (fig. 2).

Le spectromètre est constitué de trois parties:

- la tête du spectromètre contenant les détecteurs à semi-conducteurs, type Ortec; elle était placée approximativement à 50 cm de la face de la pile et entourée de plomb ( $\sim$ 10 cm);

- une canne sous vide de 2 m dans laquelle étaient tendus trois fils conducteurs reliant les sorties signal et la masse des détecteurs à la boîte de connexion;

- la boîte de connexion.



FIG.4. Spectre  $E_{\alpha} + E_t$  lissé et étalonnage du monocanal.

# 3.2. Etalonnage du spectromètre dans la colonne thermique d'HARMONIE

Nous avons étalonné préalablement le spectromètre avec la chaîne électronique associée dans la colonne thermique d'HARMONIE. Le degré de collimation était de 0,15. Les largeurs à mi-hauteur pour le dépôt de <sup>6</sup>LiF de  $30\mu g/cm^2$ , évaporé sur un des détecteurs, sont résumées dans le tableau I.

Les différences entre les résultats expérimentaux et les calculs théoriques sont également observées pour d'autres épaisseurs de dépôt de <sup>6</sup>LiF[2]. Dans le calcul théorique on a pris comme largeur à mi-



FIG.5. Réponses expérimentales pour  $E_n = 230 \pm 40$  keV (spectre de fuite).

hauteur du bruit électronique 40 keV et comme parcours des particules alpha et des tritons dans LiF respectivement 5 et 27,  $3\mu m$ .

# 3.3. Mesure du spectre $E_{\alpha} + E_t$ sans coïncidences

Les spectres  $E_{\alpha} + E_t$  ont été mesurés sans coincidences ou avec coïncidences sur une seule voie, la chaîne de spectrométrie étant incomplète à cette date pour des mesures en somme.

La première mesure  $E_{\alpha} + E_t$  a été faite sans coïncidences pour connaître l'importance du bruit de fond dans la distribution obtenue à la sortie du monocanal. Le spectre obtenu est dessiné à la figure 3 et repris après lissage à la figure 4. Nous notons un élargissement du pic, dû aux neutrons lents, de 95 à 130 keV. Malgré la couverture de plomb, le bruit de fond est encore très important. Pour  $E_n = 0$  keV le rapport du nombre d'impulsions sur le bruit de fond est de 27; à 250 keV ilest trois fois plus faible.

### 3.4. Etalonnage du monocanal

Nous branchons la connexion n° 2 (voir fig.2). Cet étalonnage a été fait avant chaque mesure de la distribution  $E_{\alpha}$  et  $E_t$  monoénergétique.

Nous avons remarqué une faible instabilité du monocanal en cours de mesure. La largeur de la fenêtre du monocanal était de 40 keV, la largeur à mi-hauteur de la distribution finale était de 80 keV avec comme énergie moyenne 230 keV.

# 3.5. Mesure des distributions $E_{\alpha}$ et $E_t$

La somme des trois distributions mesurées est représentée à la figure 5. Elle comporte trois parties:

- la distribution  $E_{\alpha}$  allant de 1, 6 MeV à approximativement 2, 5 MeV; nous pouvons facilement éliminer cette contribution dans un montage



FIG.6. Comparaison des spectres Et obtenus dans les deux détecteurs après soustraction du bruit de fond.

collimaté sans détériorer la résolution; - la distribution E<sub>t</sub> de 2,5 à 3,4 MeV;

- une distribution coïncidant avec la distribution du monocanal.

Elle contient les impulsions dues aux particules alpha et aux tritons tombant dans le même détecteur et, pour la plus grande partie, le bruit de fond. En effet, la probabilité pour que ces impulsions de bruit de fond soient formées de deux impulsions proches de 2 MeV est très faible. On le constate d'ailleurs dans la distribution mesurée par l'absence totale d'impulsions en dessous de 1,6 MeV et au-dessus de 3,4 MeV.

La figure 6 représente le spectre  $E_t$  en coincidence avec les impulsions dues aux particules alpha dans chaque détecteur, après soustraction du bruit de fond et normalisation sur  $E_t = 3$  MeV. Elle met nettement en évidence la nécessité d'une correction pour une mesure de spectre par le paramètre  $E_t$  dans le cas d'une distribution angulaire anisotrope de neutrons.

# 3.6. Mesure du spectre $E_{\alpha} + E_t$ avec des coincidences sur une voie

A la fin des mesures nous avons remesuré la distribution  $E_{\alpha} + E_t$ en faisant des coincidences sur la voie du détecteur ne comportant pas de dépôt. Notant un nouvel élargissement du pic dù aux neutrons lents (les détecteurs avaient déjà servi dans d'autres mesures) nous avons fait une deuxième mesure pour vérifier la stabilité des détecteurs. Les résultats sont reproduits à la figure 7.



FIG.7. Spectre  $E_{\alpha} + E_{t}$  en coïncidences.

TABLEAU II. RAPPORT, APRES SOUSTRACTION DU BRUIT DE FOND, DU NOMBRE D'IMPULSIONS DES SPECTRES  $E_\alpha$  +  $E_t$ OBTENUS AVEC ET SANS COINCIDENCES A DES ENERGIES DISCRETES DE NEUTRONS

			·
	E <sub>n</sub> (keV)	Rapport du nombre d'impulsions	
	100	22, 6	<u></u>
	150	20,8	
X	200	22, 9	
	250	22, 9	
	300	21, 2	
	300	23, 9	
	400	21,4	
	450	<b>24,</b> 6	
	500	27, 5	· ·
	600	22, 7	
	700	23	

Le tableau II donne le rapport du nombre d'impulsions des spectres, avec et sans coïncidences, à des énergies discrètes de neutrons et après soustraction du bruit de fond, supposé exponentiel.

L'accord observé, dans les limites des erreurs statistiques, confirme la validité des hypothèses faites pour la soustraction du bruit de fond.

# 4. INTERPRETATION DES RESULTATS

#### 4.1. Généralités

Cinq buts peuvent être recherchés dans la technique décrite.

1) Déterminer le facteur de correction à appliquer au spectre  $E_t$  ou  $E_{\alpha} + E_t$  mesuré, dû à l'anisotropie de la distribution angulaire des neutrons.

Dans ce cas la source primordiale d'erreur sera liée à la définition sur l'énergie neutronique sélectionnée.

Dans l'hypothèse d'une variation lente de la distribution angulaire et du flux de neutrons en fonction de l'énergie, le facteur de correction sera obtenu en divisant la distribution mesurée par la distribution théorique. La distribution théorique est calculée, pour la bande d'énergie neutronique analysée, sur la base des sections efficaces différentielles d'émission des tritons dans le cas d'une distribution angulaire isotrope des neutrons.

2) Déterminer la distribution angulaire des neutrons.

La précision sur la mesure sera en plus liée dans ce cas à la valeur de l'angle de collimation et à la résolution en  $E_t$  et  $E_{\alpha}$  +  $E_t$ .

En négligeant l'influence de la résolution finie et dans l'hypothèse d'une collimation parfaite des particules émises et d'une définition parfaite de l'énergie du neutron, nous pouvons écrire:

avec 
$$\frac{R[E_t(\theta, E_n)]}{S[E_t(\theta, E_n)/E_n]} = \Phi(\theta)$$

- $S(E_t/E_n)$  = réponse à un flux isotrope de neutrons monoénergétiques d'énergie  $E_n$  (déduite de la section efficace différentielle d'émission des tritons)
- $R(E_t)$  = réponse à des neutrons monoénergétiques d'énergie  $E_n$  avec une distribution angulaire quelconque du flux

 $\Phi(\theta)$  = distribution angulaire

A

= angle entre le neutron et le triton émis (lab).

La relation  $E_t = E_t(\theta, E_n)$  est donnée par la cinématique de la réaction. Si la distribution angulaire des neutrons est cylindrique et si l'axe de collimation concorde avec l'axe de symétrie, nous obtenons la distribution angulaire  $\Phi$  ( $\theta, \phi$ ). Dans le cas d'une distribution angulaire quelconque, il est nécessaire de faire tourner l'axe de collimation pour obtenir  $\Phi(\theta, \phi)$ .

3) Vérifier ou compléter les données sur les sections efficaces différentielles dans un flux isotrope de neutrons.

En effet si  $\Phi(\theta)$  = Cte nous obtenons S[ $E_t/E_n$ ], qui est reliée directement à la section efficace différentielle angulaire par la relation

$$S(E_t/E_n) = 2\pi A(E_n)\sigma_{\Omega}[\mu(E_t), E_n]$$









avec A(E<sub>n</sub>) = 2/(E<sub>t<sub>0</sub>°</sub> - E<sub>t<sub>180</sub>°) σ<sub>r</sub> = section efficace différentielle angulaire dans le système du centre de masse μ = cos⊙ = A(E<sub>n</sub>)E<sub>t</sub> - B(E<sub>n</sub>) Θ = angle d'émission du triton dans le système de centre de masse</sub>

Par suite de la résolution finie sur la distribution  $E_{\alpha}$  +  $E_t$  cette technique n'est applicable que pour des énergies neutroniques supérieures à 500 keV.

Er (MeV)	K (1 <sup>er</sup> cas)	K (2 <sup>e</sup> cas)	K (3 <sup>e</sup> cas)
0,100	0, 97	1, 11	
0,150			1, 22
0,20	1,02	0,90	0, 91
0,25			1,04
0,3	1,05	0, 93	1,01
0,35			1,14
0,4	1,07	1,03	0, 59
0,5	1,06	1, 13	
0,55			1,19
0,6	1,05	1,10	1,04
1	0,83	0,82	0, 72
2	0, 93	0, 98	1,16
		Į	

TABLEAU III. CORRECTIONS A APPORTER AUX SPECTRES  $E_{\alpha} + E_t$  MESURES, PAR SUITE DE L'ANISOTROPIE DE LA DIS-TRIBUTION ANGULAIRE DES NEUTRONS DANS TROIS CAS PRECIS

Dans les trois cas un analyseur biparamétrique permet de multiplier le nombre de réponses simultanées selon la nécessité.

4) Déterminer en fonction de l'énergie du neutron, dans le cas d'une distribution isotrope de neutrons et d'une détection  $4\pi$ , le pourcentage de particules secondaires émises faisant entre elles un angle inférieur à  $180^{\circ}$ .

5) Par une technique semblable, éliminer l'apport de neutrons au-dessus d'un certain seuil d'énergie. Ceci est important par exemple dans le cas d'un spectre très dur où les hautes énergies peuvent, par le fait que la spectrométrie  $E_t$  est une technique indirecte, noyer la réponse aux neutrons de plus faible énergie dans la distribution en énergie des tritons.

# 4.2. Interprétation de la distribution Et mesurée

Dans la figure 8 nous avons repris la distribution  $E_t$  mesurée en la comparant à la réponse théorique calculée, dans le cas d'un flux isotrope de neutrons, pour un groupe d'énergie rectangulaire de neutrons de 200 à 270 keV.

La figure 9 donne le terme de correction obtenu par division des deux distributions.

En négligeant l'erreur due à la résolution finie sur  $E_t$  et  $E_{\alpha}$  +  $E_t$  nous pouvons déduire de la distribution mesurée  $\Phi(\theta)$  en passant de la variable  $E_t$  à la variable  $\theta$ . Ceci a été fait pour  $E_n = 225$  keV et 250 keV. Les nouvelles abscisses sont reprises dans la figure 9.

Nous remarquons que la distribution est proche de l'isotropie pour  $0^{\circ} < \theta < 80^{\circ}$ , qu'elle présente un maximum autour de 100°, dû probablement à la diffusion dans le plomb, et tombe rapidement vers zéro pour des angles plus grands.

Dans le tableau III nous donnons le terme de correction K à appliquer au spectre  $E_{\alpha} + E_t$  mesuré dans les trois cas simplifiés suivants:

1)	$\Phi(\theta) = 0$	pour $110^\circ < \theta < 180^\circ$
	= 1,	5 $80^\circ < \theta < 110^\circ$
	= 1	$0^{\circ} < \theta < 80^{\circ}$
	exemple	représentatif de la mesure présente
<b>9</b> )	$\Phi(0) = 0$	00° < 0 < 1 00°

÷,	* (°) -	0	pour	00 101.	100
	=	1		$0^{\circ} < \theta <$	80°
3)	Φ(θ) = =	1 0	pour	θ = θ ≠	0° 0°

L'importance du facteur de correction dans le cas du paramètre  $E_t$  est mise en évidence par les mesures de la figure 6.

#### 4.3. Sources d'erreurs

La mesure met en évidence que les erreurs sont liées à quatre paramètres fondamentaux:

- l'épaisseur du dépôt de <sup>6</sup>LiF, qui, avec la chaîne électronique, définit la résolution que nous pouvons obtenir pour  $E_t$  et  $E_{\alpha}$  +  $E_t$ 

- le degré de collimation, qui sera, selon le but recherché. celui de la mesure du spectre  $E_t$  ou  $E_{\alpha}$  +  $E_t$  ou le plus faible possible pour la mesure de la distribution angulaire des neutrons

- la largeur de la bande d'énergie choisie, liée à la résolution de  $E_{\alpha} + E_t$ , à la définition de l'énergie moyenne dans le spectre  $E_{\alpha} + E_t$ , donc à la statistique, et à la stabilité du monocanal même

- la précision statistique.

Le rendement et les conditions expérimentales détermineront le choix optimal de chaque paramètre, et le but recherché déterminera l'importance relative donnée à chacun d'eux.

#### 5. CONCLUSIONS

Nous avons développé une nouvelle technique de mesure permettant d'interpréter d'une façon plus exacte les spectres  $E_t$  ou  $E_{\alpha} + E_t$  mesurés en pile et permettant de donner des informations sur la distribution angulaire des neutrons, en l'illustrant par un exemple. Nous avons également souligné les autres possibilités de la technique dans la mesure des spectres et de données fondamentales de la réaction <sup>6</sup>Li(n, t) <sup>4</sup>He.

### REMERCIEMENTS

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# DE LEEUW et DE LEEUW

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# DISCUSSION

D. STEGEMANN: I have two questions; firstly, what is your total spectrometer efficiency for triton collimation, as opposed to the detection of alpha particles and tritons? Secondly, what is your estimate of the accuracy obtainable with currently available  $^{6}$ Li (n,  $\alpha$ ) cross-section data?

G. de LEEUW: Where the purpose of the measurement is to correct a measured spectrum, no modification is made to the spectrometer. The total yield is then the measured yield multiplied by a factor which depends on the size of the mono-channel window. For measuring the angular distribution of neutrons, the yield depends on the definition it is desired to obtain with regard to neutron energy and the angle between neutron and triton, hence on the degree of collimation.

Adequate statistics can usually be obtained within the maximum integrated flux tolerable for the detector in question.

With regard to your second question, greater accuracy regarding total cross-sections is desirable. The data on the differential cross-sections for emission of tritons are plainly inadequate, particularly for neutron energies below 200 keV. The experimental spectrum  $E_e$  for very low energies can only be interpreted on the basis of extrapolations.

Where the sum of particle energies is being measured, the differential cross-section comes into the calculations only in a correction term, and lesser accuracy is therefore required. For the measurements which we are proposing to undertake the degree of accuracy will vary directly with the neutron energy.

# A NEW METHOD OF MEASURING ALPHA (E) FOR <sup>239</sup> Pu

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#### Abstract

A NEW METHOD OF MEASURING ALPHA(E) FOR <sup>239</sup>Pu, A special liquid scintillator detector has been developed for the purpose of measuring alpha(E) for <sup>239</sup>Pu in the energy range 10 eV to 30 keV using the Harwell Linear Accelerator time-of-flight spectrometer. (Alpha(E) is the ratio of capture to fission cross-sections as a function of incident neutron energy.) The detector has two outputs, one responding to gamma-ray interactions and the other to fast neutrons. The efficiency of the detector for gamma rays is arranged to be proportional to the gamma-ray energy. This property is achieved by utilizing an improved Moxon-Rae design and ensures that the efficiency of the detector for radiative capture events is constant irrespective of the nature of the gamma-ray cascade. The fast neutrons are also detected in the liquid scintillator and pulse shape discrimination is used to reject events produced by gamma rays. As a gamma-ray detector the device is sensitive to both radiative capture events and to the prompt gamma rays produced in fission. However, a correction for this latter component is made using the information from the fast neutron output which is essentially only sensitive to fission events. For each of the time-offlight timing channels the ratio of the corrected counts from the gamma detector to the number of fission events detected is equal to  $K \times$  alpha(E), where K is a constant determined by normalization. The technique of measuring both capture and fission simultaneously ensures that incident neutron energy spectrum charges and resolution effects are unimportant and also reduces the multiple scattering corrections. The detector system is described and some of the data obtained are shown.

#### 1. INTRODUCTION

The value of the capture cross-section  $(\sigma_n\gamma(E))$  of <sup>239</sup>Pu in the energy range 100 eV to 15 keV is one of the major uncertainties in the nuclear data required to calculate the properties of large dilute fast reactors. At the present time the errors in  $\sigma_n\gamma(E)$  in this energy range vary between  $\pm 17^{\circ}/_{\circ}$  and  $\pm 80^{\circ}/_{\circ}$  but if these can be reduced to  $\pm 5^{\circ}/_{\circ}$  then it should be possible to calculate the breeding gain to  $\pm 0.03$ . Smith [1] has stated that an error in breeding gain of  $\pm 0.1$  for a typical large sodium cooled fast reactor may result in a range of fuel doubling time of 10-20 years which could lead to variations of over 10°/o in the estimated cost of electricity generation. Therefore there are cogent reasons for making improved measurements.

In this paper a new method to measure the value of  $\sigma_{n\gamma}(E)$  or  $\alpha(E)$ (the ratio of the capture to fission cross-sections) for <sup>239</sup>Pu will be described and some provisional results presented. Before this is done, however, the data available below 10 keV will be briefly reviewed and the basic techniques of making  $\alpha$  measurements will be discussed.

### 2. REVIEW OF DATA AVAILABLE AND POSSIBLE METHODS OF MEASUREMENT

At the present time uncertainties in the values of  $\sigma_{n\gamma}(E)$  and  $\alpha(E)$ for <sup>239</sup>Pu in the energy range 100 eV to 15 keV are large, the errors varying between  $\sim 17^{\circ}/_{0}$  at 100 eV to  $\sim 80^{\circ}/_{0}$  at 10 keV [2]. Only two differential measurements of  $\alpha(E)$  have been performed in this energy

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region. Ignatev and Kirpichnikov [3] obtained data in the energy range 7 eV to 210 eV by detecting fission neutrons with fast neutron detectors and the gamma rays from both capture and fission events with sodium iodide detectors. Data below 205 eV have been obtained by Wang Yung-Chang et al. [4] using the pulsed reactor at Dubna. Their detector was a large cadmium loaded liquid scintillator in which fission events were distinguished from capture events by the technique of delayed coincidences.

The only other experiments which give direct information on a(E) are integral measurements. Examples of these are the KAPL data partially reported by Kanne et al. [5] and subsequently summarised by Sampson and Molino [6] and the experiments of Fox et al. [7]. However the integral measurements are made in broad neutron spectra and therefore can only be considered to give some indication of a in the energy region of interest. Above 20 keV, where mono-energetic neutron sources are available, good data exist for 239Pu.

There are basically three methods of determining  $\sigma_{\mathbf{n}\boldsymbol{\gamma}}(E)$  and a(E) for fissile material (neglecting chemical techniques used at thermal energies).

(a)  $\sigma_n \gamma(E)$  is derived from the measured total  $(\sigma_n T)$  and fission cross-sections  $(\sigma_n f)$  using the formula

$$\sigma_{n\gamma}(E) = \sigma_{nT}(E) - \sigma_{nf}(E) - \sigma_{nn}(E)$$
(1)

where  $\sigma_{nn}(E)$  is the scattering cross-section.

- (b)  $\sigma_{\Gamma}\gamma(E)$  is derived from measurements of the fission crosssection and a linear combination of the capture and fission cross-sections. This method is basically a measurement of  $K + R\alpha(E)$  where 0 < K < 1 and  $R \approx 1$ .
- (c)  $\sigma_{n\gamma}(E)$  and  $\sigma_{nf}(E)$  are measured directly.

The most reliable data for <sup>239</sup>Pu in the energy range 200eV to 10 keV have been obtained by the first method but the accuracy is poor because of uncertainties in the values of  $\sigma_{nn}$ .  $\sigma_{nn}$  in the energy range below 10 keV can be assumed to be the sum of the shape elastic and compound elastic cross-sections. For <sup>239</sup>Pu the shape elastic cross-section is accurately known [11] and the errors in  $\sigma_{nn}$  are predominantly due to uncertainties in the compound elastic cross-section. For <sup>235</sup>U the ratio of the compound elastic to the shape elastic crosssections is much smaller so that the uncertainties in the compound elastic cross-section are not so significant. Hence the capture cross-sections for <sup>235</sup>U determined using this method by Brooks et al. [10] and Uttley [11] are reasonably accurate.

Therefore in order to measure  $\sigma_n\gamma(E)$  and  $\alpha(E)$  for <sup>259</sup>Pu it is necessary to use the second or third methods. The third method is only possible if the fission events can be detected with 100% efficiency so that the capture gamma rays can be distinguished from those due to fission. In practice it is not possible to achieve 100% efficiency and so all measurements are essentially determinations of K + Ra(E).

Most previous measurements [12,8,4] made use of a large liquid scintillator and detected fissions by looking for the delayed pulses due to the fission neutrons which are slowed down and captured in the scintillator. This system is relatively uncomplicated above a few keV

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where mono-energetic neutron sources are available. For lower neutron energies the measurements must be made using a "white" spectrum neutron source with time-of-flight techniques and this greatly increases the complexity of the electronic equipment required to do the analysis. Therefore the only practicable methods of detecting fission events are either to use a fission chamber or to detect the fast neutrons emitted in fission. In the present experiment the latter method is used in conjunction with a Y-ray detector which detects the prompt fission and capture Y-rays. The experiment measures K + Ra(E) and is in principle similar to that of Ignatev and Kirpichnikov [3].

#### 3. THE NEW ALPHA DETECTOR

It has been seen in the previous section that the detector system adopted in the present experiment to measure  $\alpha(E)$  in the energy range below 20 keV consists of a gamma-ray detector and a neutron detector.

In order to perform any measurement of capture cross-section correctly the detector must have a constant efficiency irrespective of the form of the gamma ray cascade which follows neutron capture. This can be obtained by three methods:

- (a) using a large liquid scintillator which has an efficiency of almost 100% for any capture event.
- (b) using the system proposed by Maier-Leibnits and described by Macklin and Gibbons [13].
- (c) using a Moxon-Rae detector [14] whose efficiency is proportional to the gamma-ray energy.

It is not easy to use the first two methods in the present experiment since it would be necessary either to construct a new type of large liquid scintillator if neutrons and gamma-rays are to be detected simultaneously or to use two dimensional recording of time-of-flight and pulse height with the Maier-Leibnitz system. Therefore a detector based on the Moxon-Rae principle has been used in the present experiment.

The best method of detecting fast neutrons with good time resolution is probably to use a liquid scintillator with pulse shape discrimination to reject the pulses due to Y-rays. However when plutonium samples are used there is the added complication that the 239Pu has a decay rate of  $\sim 2.3 \times 10^9$  nuclei per second per gram and 0.06% of these disintegrations give gamma rays of energy greater than 100 keV. Furthermore, since the light output can be up to four times as great for an electron as for a proton of the same energy, the near simultaneous occurrence of two of these low energy gamma rays can easily be mistaken by the pulse shape discrimination system for a proton pulse. The classical way to eliminate this effect, which was used by Patrick et al. [15], is to put approximately 1 inch of lead between the scintillator and the sample. However it is necessary in the present experiment to detect fission and capture events simultaneously so that the corrections for multiple scattering are minimised and incident neutron spectrum changes and resolution effects are then unimportant. Therefore the presence of large amounts of lead is undesirable.

White [16] of the Electronics and Applied Physics Division at Harwell has developed a new pulse shape discrimination system which is based upon a fast linear gate and integrator. The current pulse from the anode of the detector photomultiplier is split equally between two linear



FIG.1. Sectional view of the detector used to measure alpha (E).

gates and a threshold discriminator detects the start of the pulse and opens the gates for 50 ns and 500 ns respectively. The pulses due to protons and electrons are then distinguished by checking whether or not the ratio of the integrated currents is greater than a predetermined value. The total time for analysing a pulse is approximately 1 $\mu$ s and during this period the gates are inhibited. The system is able to analyse pulses at over 100 kc/s because the integrators are restored before the inhibit is removed and the neutron output is inhibited if the fast discriminator sees a second pulse during the period when the gates are open. This system is a great improvement over the original methods of pulse shape discrimination. The system of Brooks [17], and the cross-over method, for instance, are typically limited to 3 kc/s and 20 kc/s respectively.

These developments in pulse shape discrimination techniques mean that it is now possible to combine the fission neutron and gamma ray detectors because large quantities of lead are not required. At the same time it is possible to take advantage of an improved type of Moxon-Rae detector of higher efficiency which has been developed by Weigmann et al. [18].

The new detector for the measurement of  $\alpha(E)$  is shown diagrammatically in Fig. 1. It consists of a tank of NE 213 liquid scintillator 14 inches long, 6 inches deep and 6 inches wide. In the centre 6 inches of the tank there is a 0.036 inch aluminium separator which essentially divides this region into 8 sections, these 8 sections being viewed alternately by two photomultipliers (A and B) at the ends of the tank. Gamma rays are detected by demanding a coincidence between the photomultipliers A and B; the efficiency is proportional to the gamma ray energy for effectively the same reasons as in a conventional Moxon-Rae detector. Fast neutrons are detected by separately applying pulse shape discrimination techniques to the signals occurring in the photomultipliers A and B. The proton recoils resulting from fast neutron interactions do not result in coincidences since the aluminium separator is opaque to protons with energies less than ~8 MeV; in comparison a 1 MeV electron loses ~200 keV in the aluminium. In principle errors can arise if two gamma rays are detected simultaneously in the two halves of the detector. However the efficiency of the detector for gamma rays is sufficiently small that this is not a serious effect. It is necessary of course to have relatively low efficiency detectors otherwise there would be an appreciable probability that the neutrons and gamma rays from fission would be detected in coincidence and hence cause ambiguity in the pulse shape discrimination system.

In the experimental arrangement shown in Fig. 1 a  $^{1}/4^{"}$  thick lead screen between the plutonium sample and the detector is not shown. This small amount of lead proved to be desirable to reduce the large number of small pulses in the detector due to the natural radioactivity of the sample which the pulse shape discrimination equipment has to analyse. In order to verify that the efficiency characteristics of the capture detector were not affected by the lead the detector was checked using the "black" resonance technique [14] with Ag, Pt, Au, Ta and  $^{238}$ U and also with a Monte-Carlo computer code.

#### 4. THE EXPERIMENTAL ARRANGEMENT

The new detector was assembled on one of the flight paths of the Harwell time-of-flight neutron spectrometer based on the 45 MeV Electron Linear Accelerator with its neutron 'Booster' target [19]. The main experimental details for the measurements covering the incident neutron

# TABLE I. EXPERIMENTAL DETAILS

Energy range	10 eV - 30 keV
Flight path length	34.9 m
Neutron burst width (unmoderated)	220 ns
Timing channel width	125 ns
Nominal resolution	7.2 ns/m
Resonance filters for background measurement	Mn, Mo, Ta, Al
Permanent 'black' filter	A1
Incident spectrum measurement	1/8" thick <sup>6</sup> Li glass scintillator

energy range 10 eV to 30 keV are given in Table I. Three 2 inch diameter  $^{239}$ Pu samples with low (0.70/o)  $^{240}$ Pu content and thickness of 0.00120, 0.000579 and 0.00029 atoms/barn respectively are used. The energy spectrum of the incident neutron flux is also measured with the same resolution by replacing the sample with a  $^{1}/8$  inch thick <sup>6</sup>Li glass scintillator (NE 905). The glass, which is  $2^{1}/2$  inches in diameter and is mounted  $2^{1}/16$  inches away from the photomultiplier to reduce the effect of multiple scattering, has a known efficiency as a function of neutron energy.

# 5. ANALYSIS OF DATA AND RESULTS

The number of counts from the gamma ray detector,  $N\gamma$ , and from the fission neutron detector,  $N_n$ , in a given time-of-flight channel is related to the number of capture,  $n_c$ , and fission,  $n_f$ , events by the equations

$$N_{\gamma} = \epsilon_1 n_{f} + \epsilon_2 n_{c} \tag{2}$$

$$N_{n} = \epsilon_{3} n_{f} + \epsilon_{L} n_{c}$$
(3)

where  $\epsilon_1$ ,  $\epsilon_2$ ,  $\epsilon_3$  and  $\epsilon_4$  are the efficiencies of the two detectors for fission and capture events respectively. The values of  $n_c$  and  $n_f$  depend upon the capture and fission cross-sections through the following relationships

$$n_{c} = \phi(1-T) \left\{ \frac{\sigma_{nY}}{\sigma_{nT}} + \frac{\sigma_{nn}}{\sigma_{nT}} < (1-T') \frac{\sigma_{nY}}{\sigma_{nT}'} > + \cdots \right\}$$
(4)  
$$n_{f} = \phi(1-T) \left\{ \frac{\sigma_{nf}}{\sigma_{nT}} + \frac{\sigma_{nn}}{\sigma_{nT}} < (1-T') \frac{\sigma_{nf}}{\sigma_{nT}'} > + \cdots \right\}$$
(5)

where  $\beta$  is the incident neutron flux,  $\sigma_{nT}$ ,  $\sigma_{n\gamma}$ ,  $\sigma_{nf}$  and  $\sigma_{nn}$  are the total, capture, fission and scattering cross-sections and T is the transmission of the sample. The triangular brackets denote averages and represent the probability of a scattered neutron causing a further

reaction; the primed quantities are the values of the cross-sections etc. after scattering.

The equations are simplifications because they do not allow for the 0.005" aluminium can round the sample or the small  $(1.07^{\circ}/^{\circ})$ fraction of aluminium in the sample. The more complete equations will be used in the final analysis of the data but these simplified equations can be used to demonstrate that in an alpha measurement of this type the multiple scattering corrections are not very significant. If equation (4) is divided by equation (5) and the result simplified we obtain

$$\frac{\mathbf{n}_{c}}{\mathbf{n}_{f}} = \alpha \quad \left\{ \frac{1 + \sigma_{nn} \left\langle \frac{(1 - \mathbf{T}')}{\sigma_{nT}'} - \frac{\alpha'}{\alpha} - \frac{\sigma_{nf}'}{\sigma_{nf}} \right\rangle + \dots}{\left\{ 1 + \sigma_{nn} \left\langle \frac{(1 - \mathbf{T}')}{\sigma_{nT}'} - \frac{\sigma_{nf}'}{\sigma_{nf}} \right\rangle + \dots} \right\}}$$
(6)

where  $\alpha = \frac{\sigma_n \gamma}{\sigma_n \rho}$  and  $\alpha' = \frac{\sigma_n \gamma'}{\sigma_n \rho'}$ 

Hence if a' = a then  $\frac{n_c}{n_f}$  = a. In practice a is a function of neutron energy but in general the variations are much analyzed. variations in cross-sections. Therefore, since the samples are thin,

the ratio 
$$\frac{n_c}{n_p}$$
 is expected to be nearly equal to a.

Hence the value of a can be obtained by simplifying equations (2) and (3) to give

 $\frac{n_{c}}{n_{f}} = \alpha S = \frac{\frac{A}{N_{T}} \frac{N_{Y}}{N_{n}} - 1}{\frac{B - C}{N_{T}}}$ (7)

where S is the multiple scattering correction and

$$A = \frac{\epsilon_{3}}{\epsilon_{1}}, B = \frac{\epsilon_{2}}{\epsilon_{1}}, C = \frac{\epsilon_{4}}{\epsilon_{1}}.$$

In this experiment it is being assumed that all the efficiencies  $(\epsilon_1, \epsilon_2, \epsilon_3, \epsilon_4)$  are independent of incident neutron energy. This assumption is undoubtedly correct for  $\epsilon_2$  because of the properties of a Moxon-Rae detector and because the total energy of capture gamma-rays (~6 MeV) is large compared with the incident neutron energy.  $\epsilon_{4}$  is small and so any changes in the capture gamma-ray spectrum as a function of incident neutron energy can be neglected.

However, the values of  $\epsilon_3$  and  $\epsilon_1$ , which are the efficiencies of the neutron and gamma-ray detectors for fission, could change because the fission mass distribution and the fission fragment kinetic energy have been observed to change from resonance to resonance. Since the detector system only gives one pulse per fission event the efficiency €3 is given by

$$\varepsilon_{3} = \xi \left(1 - \left(1 - \varepsilon_{n}\right)^{\vee}\right) P(v)$$
(8)



FIG.2. Neutron yield from 0.00058 atoms/barn <sup>239</sup>Pu sample.

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FIG. 3. Gamma-ray yield from 0.00058 atoms/bam <sup>239</sup>Pu sample.







FIG.5.  $\alpha$  (E) for <sup>239</sup> Pu.

where  $P(\mathcal{V})$  is the probability of emitting  $\mathcal{V}$  neutrons per fission and  $\epsilon_n$  is the efficiency of neutron detector for neutrons. Bollinger et al. [20] have made a deliberate attempt to examine <sup>239</sup>Pu for variations in the average number of neutrons per fission ( $\overline{\mathcal{V}}$ ) from resonance to resonance. They found that within the statistical accuracy of 3-4/o there was no perceptible variation. Recently Cao et al. [21] have measured the fission cross-section of <sup>235</sup>U using simultaneously the fragment and neutron detection methods. They conclude that the data obtained agree within a few per cent, any deviations being readily explicable as due to statistics and self absorption and multiple scattering effects. These two pieces of information support the assumption that  $\overline{\mathcal{V}}$  and  $\epsilon_3$  are constant from resonance to resonance.

The changes in the relative fission gamma ray yield as a function of fragment mass have been measured [22] and it is found that these variations are about 50% of those occurring in  $\Im$  [23]. Therefore, since  $\Im$  can be taken to be constant, it can be assumed that the relative Y-ray yield and hence &1 are constant.

At the present time measurements of the neutron and  $\gamma$ -ray yield have been made for the two thickest <sup>239</sup>Pu samples, the incident neutron flux has been measured and the runs on Ag, Pt, Au, Ta and <sup>238</sup>U required to check the performance of the  $\gamma$ -ray detector have been made. Figs. 2 and 3 show the observed neutron and gamma yields in the energy range 11 eV to 100 eV from the sample of medium thickness. The data are untreated except for background subtraction and the figures on the graphs give the resonance energies.

The experiment is normalised by assuming the values of a for 13 well resolved low energy resonances in the energy region below 80 eV. Fig. 4 shows the values of a and the corresponding values of  $N_{\gamma}/N_{n}$  for the medium sample. The curve is the best line fitted to the data which assuming that the multiple scattering correction S is unity leads to values of A = 0.930, B = 0.390 and C = 0.00683. The values of a for the resonances were obtained from the data of Derrien et al. [24].

The reactor physicist requires the ratio of the average capture cross-section to the average fission cross-section  $(\langle \sigma_{\Pi \gamma} \rangle / \langle \sigma_{\Pi f} \rangle)$  rather than the average value of a  $(\langle a \rangle)$ . In order to get the average value of these cross-sections it is necessary to make multiple scattering corrections and the present experiment is not far enough advanced for this to be done. However, in order to see whether or not the value of  $\langle \sigma_{nY} \rangle / \langle \sigma_{nf} \rangle$  is high in the energy range from a few hundred eV to 10 keV the neutron and Y-ray counts have been integrated over 100 eV intervals below 1 keV, 1 keV intervals between 1 and 10 keV, 5 keV intervals from 10 to 30 keV and 10 keV intervals from 50 to 70 keV. Then, using equation (7) an estimate of  $\langle \sigma_n \gamma \rangle / \langle \sigma_n f \rangle$  can be made. The results are shown in Fig. 5 and it can be seen that the results are in reasonable agreement with the values obtained from  $\sigma_{nT}$  and  $\sigma_{nf}$ . At the high energy end between 10 and 30 keV the results are higher than expected from the data of De Saussure et al. but this discrepancy is not considered to be significant at this stage. The errors shown in Fig. 5 have been arbitrarily assumed to be  $\pm 33^{\circ}/o$  because systematic errors, which have not been assessed, predominate in this experiment. The procedure used to obtain some indication of the value of  $\langle \sigma_{nY} \rangle / \langle \sigma_{nf} \rangle$  is only valid if the multiple scattering corrections are small and the variation of the incident neutron flux over the energy interval used is negligible. In addition corrections are required for the effect of the0.005"Al can round the samples. Therefore the results shown in Fig. 5 should be viewed with extreme caution. However, the results do show, particularly in the energy range above 1 keV where the assumptions are not too unreasonable, that the value of a is probably of the order of unity and not of the order of 0.5 as suggested by the KAPL data [6].

#### 6. ACKNOWLEDGMENTS

The authors are particularly indebted to Mr. G. White for his co-operation and development of the pulse shape discrimination equipment, to Mr. M. J. Hopper for converting his Y-ray Monte Carlo programme for use with the present detector system and to Mr. M. C. Moxon for advice and discussions on capture cross-section measurements. The advice and encouragement of Dr. E. R. Rae is gratefully acknowledged.

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### DISCUSSION

H.W. KÜSTERS: The  $\alpha$  curve for <sup>239</sup> Pu used in evaluating the capture data in the SNEAK set presented in paper SM-101/12 is based on extrapolated Hopkins-Diven data below 20 keV, following the KAPL foil

measurements. This  $\alpha$  curve is in good agreement with the Douglas-Barry evaluation, but it turns out both from experiment and theoretical investigations that the  $\alpha$  values below 20 keV are higher than those we used by up to 30%. This is bound to have a considerable influence on the neutron characteristics of large plutonium-fuelled fast reactors, especially for steam-cooled systems, where the breeding gain will be even lower than it is now.

Have your data been included in a group cross-section set and have the effects referred to already been calculated?

M.G. SCHOMBERG: These data are not included in the FD2 crosssection set but the effect has been tested in modified versions of this set. The new FD3 set will contain high  $\alpha$  values in line with these recent values obtained at Harwell.

The data at present are provisional and it is difficult to recommend the best  $\alpha$  values to use, particularly in the 1-15 keV range. The values quoted in my paper would result in a reduction in the breeding gain for a typical large sodium-cooled fast reactor of about 0.1, as compared to the value calculated with FD2; however, with suitable design a good doubling time may still be achieved. The effect on the steam-cooled reactor has not been studied in detail but it is expected to be more serious, especially in view of the low breeding gain calculated using the older data.

J.Y. BARRÉ: I should like to mention that with the fitting method described in paper SM-101/58 we expect to increase the value of the parameter  $\alpha$  for <sup>239</sup>Pu between 10 and 100 keV by about 15%. This would tally with your latest measurements.

K.H. BECKURTS: What is the sensitivity E of your system for capture and fission events? Also, what is the time resolution of your system, and do you plan to use it for 'fast timing'?

M.G. SCHOMBERG: The measured efficiency for one detector, as used in the system described, is of the order of 5% for both capture and fission events.

The timing accuracy of both the neutron and gamma detectors depends mainly on the choice of photomultipliers. With fast photomultipliers a timing resolution of less than 2 nsec has been obtained and the system has been used for 'fast' time-of-flight measurements.

# GENERAL DISCUSSION

F. EBERSOLDT: I should like to make a few remarks on our method of determining neutron spectra from reaction rates and a hypothetical spectrum.

The general equation relating the neutron flux  $\phi(E)$  to the response function of a detection system R(E) is an integral Fredholm equation of the first kind. The kernel of this equation contains essentially the crosssections of the reactions considered.

When measuring reaction rates (spectral indices, for instance) to determine neutron spectra, one no longer obtains a response function continuously dependent on a parameter E (as in proton recoil spectrometry, for example), but only a few discrete values. Their number N, which is equal to the number of detectors, is unfortunately not very high. Furthermore, the measure points  $r_k$  are affected by statistical errors  $\epsilon_k(k=1, \ldots, N)$ 



One could try a direct inversion of Eqs(1) using a multigroup technique, for instance. But the cross-section matrix appearing in these equations would often be unfavourable because of the similarity of some cross-sections. This would produce solutions which are very sensitive to errors in the reaction rates or cross-sections and, as stated by several authors, show unwanted and non-physical oscillations. All this excludes a direct inversion of the fundamental equations (1).

There have been two different methods used mainly up to now to solve the equations under consideration. The first one assumes a priori a special shape of the spectrum. In a parametric representation of the spectrum the parameter values are determined from the measured reaction rates. The difficulty of this method consists in choosing good approximation functions and their domain of definition.

Another way to solve the problem is to develop the function  $\phi$  (E) on a basis of orthogonal functions and to determine the coefficients of the development from the measured reaction rates. This method has the disadvantage of giving solutions which may oscillate because of the limited number of terms in the development.

The method presented here uses the fact that, besides the experimental quantities  $r_k$ , one generally has a rough idea of the spectrum sought. This can be gained from calculations or similar experiments made previously. The principle of the technique is to start with this hypothetical spectral function  $\varphi_0$  (E) and to add a correction function  $\varphi_1$ (E) which is to be determined from the measure points

$$\phi(\mathbf{E}) = \mathbf{x} \cdot \varphi_0(\mathbf{E}) + \varphi_1(\mathbf{E}) \tag{2}$$

(1)

The quantity x is an amplitude factor which is unknown at the beginning. It will be chosen so as to minimize the total correction, i.e.

$$\int_{0}^{\infty} \left[ \varphi_{1}(\mathbf{E}) \right]^{2} d\mathbf{E} \to \text{minimum}$$
(3)

As one assumes that  $\varphi_0(\mathbf{E})$  is a 'physical' function, i.e. a continuous one with first derivative also continuous, one should require these properties to hold for the correction function  $\varphi_1(\mathbf{E})$  too.

It seems even realistic to require that  $\varphi_1(E)$  should be a 'smooth' curve, in particular without strong oscillations. This can be realized by minimization of the curvature, i.e.

$$\int_{0}^{\infty} \left(\frac{d^2 \varphi_1}{dE^2}\right)^2 dE \rightarrow \text{minimum}$$
 (4)

These physical requirements lead, of course, to an approximate solution of the equations (1), with some contributions to the error functions  $\epsilon_k$ . But one should keep in mind that the exact solution generally would not have any physical meaning because of its undesirable oscillations.

Introducing the multigroup formalism to solve the problem, one can use the technique of Lagrange's multiplier to combine Eq. (4) with the condition that the total error is a constant

$$\sum_{k=1}^{N} \epsilon_{k}^{2} = c^{2} \qquad (c = const) \qquad (5)$$

$$\lambda \sum_{i=1}^{N} \left( \varphi_{1}^{i+1} - 2 \varphi_{1}^{i} + \varphi_{1}^{i-1} \right)^{2} + \sum_{i=1}^{N} \epsilon_{i}^{2} - c^{2} \rightarrow \min$$
 (6)

where  $\lambda$  is the inverse of the Lagrange multiplier.

As opposed to the classical theory where  $\lambda$  is calculated, it seems better to attribute a non-negative value to  $\lambda$  and to omit the constant term -c<sup>2</sup> which is often unknown and can be calculated later by Eq. (5).

From the Lagrange equations (6) and the multigroup form of equations (1), one obtains the final results

$$\vec{\epsilon} = -\lambda \, \mathrm{A} \, \vec{\phi}_{1}; \, \vec{\phi}_{1} = (\mathrm{F} + \lambda \, \mathrm{A})^{-1} \, (\vec{\mathrm{r}} - \mathrm{x}_{0} \cdot \vec{\mathrm{s}}) \tag{7}$$

with

$$\mathbf{x}_{0} = \frac{\left(\left(\mathbf{F} + \lambda \mathbf{A}\right)^{-1} \vec{\mathbf{s}}, \left(\mathbf{F} + \lambda \mathbf{A}\right)^{-1} \vec{\mathbf{r}}\right)}{\left(\left(\mathbf{F} + \lambda \mathbf{A}\right)^{-1} \vec{\mathbf{s}}, \left(\mathbf{F} + \lambda \mathbf{A}\right)^{-1} \vec{\mathbf{s}}\right)}$$
(8)

The matrices A and F contain only average values of cross-sections. The components of the vector  $\vec{s}$  are obtained by the scalar products of  $\varphi_0(E)$  with all the cross-sections  $\sigma_k(E)$ .

The parameter  $\lambda$  controls the rate of smoothing of  $\varphi_1(E)$ . Starting with  $\lambda=0$ , which gives the direct unfolding of equations (1) without smoothing (correct solution), one increases the amount of smoothing when increasing  $\lambda$ .

The best  $\lambda$  seems to be one that eliminates undesirable oscillations in the solution to  $\varphi_1(E)$  and gives at the same time an error  $|\epsilon_k|$  exceeding the experimental error, which of course is a lower limit for  $|\epsilon_k|$ .

The method has been programmed in FORTRAN IV for the IBM-360 computer, and the code is still being tested. The first applications to mathematical models, where the theoretical solutions were known, have given very satisfactory results.

The advantages of this method are the following:

(1) The solutions obtained are 'physical' functions without errorinduced oscillations

(2) No spectral shape is fixed a priori

(3) The solutions seem to be little sensitive to the choice of the initial function  $\varphi_0$  (E), thus all the necessary importance is given to the measured reaction rates.

(4) Furthermore one gets a measure of the overall error affecting the measure points.<sup>1</sup>

H.W. KÜSTERS: As I see it, you are trying to adjust the spectrum to integral data. I would have thought that there was no difficulty in predicting the spectrum if one really has the correct cross-section data. Could you clarify whether in your evaluation you kept constant the effective cross-sections, which are the really uncertain data?

F. EBERSOLDT: Spectra calculated by multigroup calculations, for instance, do not necessarily agree with experimental results, essentially because the cross-sections used are not sufficiently good. The method presented here allows us to correct these calculated spectra by reaction-rate measurements (spectral indices). Errors in the cross-sections used for the theoretical determination of the spectrum contribute to the error  $\epsilon$  affecting the measured reaction rates. To prevent these errors  $\epsilon$  having too great an influence on the calculated spectrum the matrix F describing the cross-sections of all the detectors should not be too singular. This could be arranged by choosing a good set of detectors with dissimilar cross-sections over appropriately chosen energy ranges.

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<sup>&</sup>lt;sup>1</sup> The subject of this presentation will be published more completely in a nuclear journal.

ZONED SYSTEMS (Session IV) Chairman: M. BOGAARDT

# ETUDE EXPERIMENTALE DES CARACTERISTIQUES NEUTRONIQUES DES MILIEUX MULTIPLICATEURS RAPIDES DANS L'ASSEMBLAGE CRITIQUE THERMIQUE-RAPIDE ERMINE

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#### Abstract — Résumé

EXPERIMENTAL STUDY OF THE NEUTRON CHARACTERISTICS OF FAST CORES IN THE ERMINE THERMAL-FAST CRITICAL ASSEMBLY. A section of fast-reactor lattice was placed in the central hole of the MINERVE reactor, thus forming a coupled "thermal-fast" critical assembly. The authors describe the construction of the first core, composed of the MASURCA 1-B lattice of 30% enriched uranium diluted in graphite, together with the experimental techniques used.

Most measurements were carried out by the oscillation method, using an automatic regulating rod, to compensate for reactivity effects. The experiment was conducted with special care, so as to avoid electronic, mechanical and neutron perturbations as much as possible.

The paper shows the results of measurements of spectral indices and of reactivity effects of fissile, fertile and structural materials. It also contains the results of experiments carried out to determine heterogeneity effects, the Doppler effect of <sup>238</sup> U and the importance functions.

These experimental values are compared with values calculated from transport theory. The authors analyse sources of error, various causes of perturbations and improvements required in the methods of interpretation.

The authors conclude that the difficulties met in the interpretation of these experiments are due to the fact that the core has two zones and that they are the same, whether the driver zone is thermal or fast, when the asymptotic spectrum is reached.

ETUDE EXPERIMENTALE DES CARACTERISTIQUES NEUTRONIQUES DES MILIEUX MULTIPLICATEURS RAPIDES DANS L'ASSEMBLAGE CRITIQUE THERMIQUE-RAPIDE ERMINE. Une portion de réseau de réacteur à neutrons rapides est placée dans la cavité de la pile MINERVE, formant ainsi un assemblage critique couplé «thermique-rapide». La réalisation du premier cœur, constitué par le réseau MASURCA 1-B en uranium enrichi à 30% dilué dans le graphite, est décrite ainsi que les techniques expérimentales utilisées.

La majeure partie des mesures sont effectuées par la méthode d'oscillation, les effets en réactivité étant compensés par une barre de pilotage automatique. Un soin particulier est apporté à la réalisation expérimentale afin d'éviter au maximum les perturbations électroniques, mécaniques et neutroniques.

Les auteurs présentent les résultats des mesures d'indices de spectre et d'effets en réactivité des matériaux fissiles, fertiles et de structure, ainsi que les résultats des expériences effectuées pour déterminer les effets d'hétérogénéité, l'effet Doppler de l'uranium-238 et le rapport des importances. Ces valeurs expérimentales sont comparées aux valeurs calculées par la théorie du transport. Les sources d'erreurs, les différentes causes de perturbation et les améliorations à apporter aux méthodes d'interprétation sont analysées.

Les auteurs concluent enfin que les difficultés rencontrées dans l'interprétation de ces expériences sont dues au fait que le cœur est à deux zones et qu'elles sont les mêmes, que la zone nourricière soit thermique ou rapide, lorsque le spectre asymptotique est atteint.

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# INTRODUCTION

Le développement des études sur les réacteurs rapides a entraîné la construction d'ensembles critiques de grandes dimensions qui immobilisent des quantités importantes de matières fissiles et qui souvent ne sont pas assez souples pour s'adapter aux conditions expérimentales particulières nécessitées par la mise en œuvre de techniques expérimentales précises.

Les cœurs à deux zones se sont développés récemment pour effectuer les mesures sur une partie seulement du réseau, afin de diminuer la quantité de combustible à étudier; cependant, le grand volume de la zone nourricière rapide reste un investissement important en combustible et rend l'exploitation du réacteur plus complexe.

L'expérience ERMINE est un assemblage critique à deux zones. La zone nourricière est thermique, ce qui permet de rendre l'ensemble critique avec une faible quantité de <sup>235</sup> U et, tout en maintenant la sûreté de fonctionnement des assemblages thermiques, d'atteindre au centre de la zone rapide un spectre caractéristique du milieu à étudier.

Ces expériences sont effectuées dans le cadre des études entreprises en France pour compléter les informations obtenues sur l'assemblage critique MASURCA [1] auquel sont associés, d'une part, l'expérience exponentielle HUG montée sur le réacteur HARMONIE, d'autre part, l'assemblage thermique-rapide ERMINE installé dans le cœur de MINERVE. Deux réseaux de base seront étudiés sur les trois ensembles afin d'assurer les recoupements nécessaires.

Dans ce mémoire on développe les expériences effectuées sur le premier réseau ERMINE, les techniques expérimentales utilisées ainsi que les résultats obtenus. Les conclusions sur l'interprétation des mesures sur les assemblages à deux zones thermique-rapide sont comparées à celles déduites des expériences étrangères sur les cœurs à deux zones rapides.

# 1. DESCRIPTION DE L'ASSEMBLAGE

L'assemblage thermique-rapide ERMINE se trouve dans la pile MINERVE située au Centre d'études nucléaires de Fontenay-aux-Roses. Il s'agit d'une pile piscine dont le combustible est de l'uranium enrichi à 90%. Elle possède une cavité centrale de section carrée dont le côté peut varier de façon continue, dans laquelle on a placé la partie rapide de l'assemblage ainsi que la zone d'adaptation et les réflecteurs axiaux suivant une configuration représentée sur les figures 1 et 2.

Un cube de graphite de 310 mm d'arête est percé de 108 trous contenant les éléments de matière fissile constitués de barreaux cylindriques d'uranium métallique enrichi à 30% de 305 mm de longueur et de 12,7 mm de diamètre, gainés d'acier inoxydable. Le vide correspondant à la composition normale du cœur MASURCA 1-B est ménagé entre la gaine et le graphite. Au centre du bloc de graphite on dispose d'un canal de mesures de section carrée dont les dimensions intérieures (28,  $1 \times 28$ , 1 mm) sont identiques à celles d'une cellule normale du réseau (fig. 3). La masse de <sup>235</sup>U dans la zone rapide est de 22 kg environ.

Afin d'obtenir au centre de l'assemblage le spectre caractéristique du réseau étudié avec un volume minimal, on a placé contre les faces de la zone rapide un filtre convertisseur en uranium naturel métallique, de 4 cm d'épaisseur sur les faces latérales et de 8 cm plus 7 cm d'acier inoxydable sur les faces supérieures et inférieures.

La zone nourricière thermique est formée par les éléments combustibles de MINERVE entourés des réflecteurs latéraux en graphite de 50 cm d'épaisseur, l'ensemble étant plongé dans l'eau. La masse de <sup>235</sup>U dans cette zone est de 5, 1 kg.

L'assemblage ainsi réalisé présente, du point de vue du contrôle, les mêmes caractéristiques qu'un réacteur thermique. Une étude des paramètres de couplage suivant la formulation d'Avery a montré que le coefficient de multiplication de la zone rapide était faible (il ne s'y produit qu'environ 10% des fissions) et que le temps de vie global était très proche de celui de la zone nourricière.

La zone rapide a la composition du cœur MASURCA 1-B qui est résumée dans le tableau I pour les différents milieux.

# 2. TECHNIQUES EXPERIMENTALES

Les expériences avaient pour but

- de vérifier qu'au centre de l'assemblage ainsi réalisé on obtenait un spectre caractéristique du réseau à étudier,

- de développer les techniques expérimentales en vue de leur application sur MASURCA,

- enfin d'apporter des résultats en vue de l'étude de ce premier réseau. Pour contrôler la validité des résultats, il faut s'assurer de

l'identité des effets en réactivité normalisés et des spectres directs avec ceux mesurés au centre d'un cœur critique de même composition. Au stade actuel de ces études, le recoupement complet n'est pas encore possible avec MASURCA, qui est chargé avec un cœur au plutonium, aussi la comparaison n'a-t-elle porté que sur les spectres directs mesurés à l'aide de taux de réaction sur l'empilement exponentiel HUG dont le volume dépasse la moitié du volume critique.

# 2.1. Mesures de taux de réaction

On peut distinguer

- les mesures relatives pour contrôler le domaine de stabilité du spectre au centre de l'assemblage et son identité avec le centre du massif exponentiel HUG,

- les distributions radiales des taux de fission dans la cellule centrale pour mettre en évidence les effets d'hétérogénéité,

- les mesures absolues pour comparer un certain nombre d'indices de spectre aux résultats des calculs correspondants.

Si les deux premiers types de mesures présentent peu de difficultés, le dernier pose des problèmes d'étalonnage qui ne sont pas encore entièrement résolus dans la mesure où l'on désire atteindre une très bonne précision. On a utilisé deux techniques: les chambres à fission et l'activation de détecteurs.



FIG. 3. Canal d'oscillation.

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Corps	Cœur rapide	Zone de transition	Zone nourricière	Réflecteurs
Uranium-235	$0,232 \cdot 10^{-2}$	0,303 ° 10 <sup>-3</sup>	0,134 · 10 <sup>-3</sup>	-
Uranium-238	0,536 10-2	0,0419	0,147 • 10-*	-
Acier	0,624 ° 10 <sup>-2</sup>	0,961 • 10-2	-	-
Aluminium	-	-	0,0235	0,406 ' 10-2
Graphite	0,0542	-	-	0,077
Eau	-	-	0,0196	0,100 · 10 <sup>-2</sup>

# TABLEAU I. COMPOSITION DE LA ZONE RAPIDE $(10^{24} \text{ atomes/cm}^3)$

#### 2.1.1. Chambres à fission

L'amélioration des techniques dans ce domaine a permis d'utiliser des chambres cylindriques de 4 mm et de 1,5 mm de diamètre, ces dernières étant réalisées pour des mesures de distribution fine de taux de fission dans la cellule. L'épaisseur des dépôts varie de 100 à 1000 $\mu$ g suivant les dimensions de la chambre et la nature des corps fissiles, qui sont les suivants: <sup>235</sup>U, <sup>238</sup>U,Unat., <sup>239</sup>Pu,<sup>232</sup>Th et <sup>237</sup>Np. Les impulsions délivrées par ces chambres sont amplifiées et analysées par un sélecteur d'amplitudes.

Actuellement, les seules mesures absolues concernent le rapport  $\hat{\sigma}_{f\,U238}/\hat{\sigma}_{f\,U235}$ . Elles sont effectuées à l'aide de deux chambres à fission possédant, l'une un dépôt de <sup>235</sup>U, l'autre un dépôt d'uranium naturel. L'étalonnage est effectué dans un spectre thermique qui permet d'atteindre le rapport des masses de <sup>235</sup>U dans les deux chambres. Cette méthode sera étendue aux autres couples suivants: <sup>240</sup> Pu - <sup>239</sup> Pu; <sup>237</sup> Np - <sup>239</sup> Pu; <sup>238</sup> U - <sup>239</sup> Pu; <sup>238</sup> U - <sup>233</sup> U. Il faut dans ces cas, en plus de l'étalonnage dans un spectre thermique, effectuer une analyse isotopique ou un dosage spectrométrique précis pour déterminer la quantité relative du corps fissile à basse énergie par rapport au corps dont la réaction de fission présente un seuil.

# 2.1.2. Détecteurs

Les taux de fission dans le <sup>238</sup> U et le <sup>235</sup> U sont mesurés avec des détecteurs pour déterminer

- leurs valeurs relatives dans l'uranium et le graphite,

- la valeur absolue du rapport  $\hat{\sigma}_{f\,U235}$  / $\hat{\sigma}_{f\,U235}$  dans l'uranium et le graphite pour la comparaison entre ERMINE et l'empilement exponentiel HUG.

Pour ce dernier cas, des précautions particulières quant aux choix des temps d'irradiation et de décroissance, des seuils de discrimination et des activités des détecteurs doivent être prises pour s'affranchir des erreurs systématiques. Le bon accord avec le même indice mesuré avec les chambres à fission montre que la technique adoptée est satisfaisante. La détermination de la valeur relative du taux de capture du <sup>238</sup> U de deux détecteurs se fait en mesurant, soit l'activité  $\gamma$  du pic à 106 keV du <sup>239</sup> Np après correction des produits de fission parasites, soit, dans le cas des mesures de l'effet Doppler, l'activité  $\gamma$  du pic à 74 keV du <sup>239</sup> U.

D'autres types de détecteurs, en particulier le soufre, le sodium et le manganèse, ont été utilisés pour la comparaison entre ERMINE et HUG et pour des mesures relatives entre différents points.

## 2.2. Mesure des effets en réactivité

Les effets en réactivité sont mesurés par la méthode d'oscillation. Le canal de mesure, situé au centre du réseau, a les dimensions de la cellule centrale. L'échantillon, de 101,6 mm de longueur, est placé dans un train continu de graphite et d'uranium enrichi à 30%, de façon à reconstituer la cellule; sa longueur est telle que, pendant le mouvement d'oscillation, le réseau soit toujours complet afin de ne créer que la perturbation due à l'échantillon sans modifier l'environnement (fig. 2 et 3). Cependant, pour certains échantillons de plus grand volume, le train d'oscillation est chargé uniquement de graphite.

L'oscillation est du type «pseudo-carré» avec une période de 20 s et des temps de transit de l'ordre du dixième du temps d'arrêt. L'effet en réactivité produit par le déplacement de l'échantillon est compensé par un pilote automatique constitué d'une barre rotative recouverte de secteurs de cadmium dont le mouvement est asservi aux variations du signal délivré par une chambre d'ionisation placée dans le réflecteur de la zone thermique (fig. 1). Cette barre compense automatiquement les variations de réactivité créées par l'échantillon et son angle de rotation est enregistré par un analyseur en temps, ce qui permet d'éliminer les périodes de transit dans le dépouillement des résultats. La linéarité de la barre de pilotage a été vérifiée pour la gamme d'utilisation qui représente environ  $\pm 10$  pcm. La précision atteinte avec ce dispositif de mesure est inférieure à 0, 01 pcm soit 10<sup>-7</sup> en réactivité.

#### 2.3. Effet Doppler

Une première série de mesures a été entreprise pour atteindre l'effet Doppler du <sup>238</sup> U. Deux méthodes ont été utilisées, l'une par l'oscillation d'un échantillon chauffé, l'autre par l'activation de détecteurs placés dans l'échantillon chauffé.

Pour les mesures par oscillation, les échantillons prégainés en acier inoxydable et placés à l'intérieur du train d'oscillation (fig. 2) sont chauffés par induction à haute fréquence dans le champ d'induction d'un solénoide de cuivre situé au-dessus du cœur, à une distance du milieu du cœur égale à la course d'oscillation. Pendant chaque période d'oscillation, l'échantillon chaud et un échantillon froid identique séjournent alternativement pendant une demi-période de 10 s au milieu du cœur, le four à induction ne fonctionnant que pendant la demi-période où l'échantillon chaud est placé à l'intérieur. Ce dispositif original permet, le régime d'équilibre étant établi, d'obtenir des températures parfaitement reproductibles, de changer facilement les dimensions des échantillons et de n'apporter aucune perturbation par le dispositif de chauffage dans le cœur. Les mesures ont été effectuées jusqu'à 500°C sur des échantillons d'uranium naturel métallique et d'oxyde, et sur un échantillon d'uranium métallique enrichi à 30%.

Pour les mesures par activation, deux détecteurs d'uranium appauvri sont placés respectivement au centre des échantillons chaud et froid du montage précédent. Pendant l'oscillation, ils sont ainsi irradiés pendant des temps égaux, et la comparaison des taux de comptage de la raie  $\gamma$ de 74 keV du <sup>239</sup> U formé permet d'obtenir la variation relative de la capture du <sup>238</sup> U.

# 2.4. Mesures d'importance

Les mesures relatives d'importance à l'aide de sources de neutrons sont effectuées dans les mêmes conditions que les mesures d'effets en réactivité. La source, placée dans un train continu de graphite, est oscillée entre le cœur et l'extérieur. Pour séparer l'effet sur la réactivité des neutrons émis de celui dû aux matériaux constitutifs de la source, on effectue la mesure à différentes puissances. On a aussi vérifié que cette séparation donnait de bons résultats en oscillant une source de Sb-Be vierge identique à la source active. Aucune correction n'est à apporter à la cinétique de la pile, celle-ci étant dans tous les cas maintenue à l'état critique grâce à la barre de pilotage automatique.

### 3. RESULTATS ET COMPARAISONS AVEC LES CALCULS

# 3.1. Indices de spectre

Pour la comparaison entre ERMINE et l'empilement exponentiel HUG, les indices de spectre  $\sigma_{f U238}/\sigma_{f U235}$  sont mesurés avec des couples de détecteurs de 9 mm de diamètre placés au centre de l'élément combustible de 12,7 mm de diamètre et dans le graphite au voisinage du barreau. On constate un excellent accord sur cet indice de spectre, qui est aussi confirmé pour des couples de détecteurs Mn-S et Na-S:

	ERMINE	HUG			
U <b>raniu</b> m	$342 \pm 2, 5 \cdot 10^{-4}$	$341 \pm 4, 5 \cdot 10^{-4}$			
Graphite	$288 \pm 4 \cdot 10^{-4}$	$292 \pm 2,5 \cdot 10^{-4}$			

Les mesures effectuées avec des chambres à fission miniatures permettent d'obtenir la distribution radiale des taux de fission  $R_{f\ U235}$ et  $R_{f\ U238}$  du <sup>235</sup>U, du <sup>238</sup>U et de l'indice de spectre  $\hat{\sigma}_{f\ U238}/\hat{\sigma}_{f\ U235}$ dans le graphite. Les résultats sont présentés sur les figures 4 et 5 et montrent que le taux de fission dans le <sup>238</sup>U et l'indice de spectre varient de près de 20% entre le centre du barreau et le point situé dans le graphite à la limite de la cellule.

On caractérise le milieu étudié par l'indice de spectre  $\partial_{f U238}/\partial_{f U235}$ en trois points de la cellule situés respectivement au centre de l'uranium, dans le graphite au voisinage du barreau et dans le graphite à la limite de la cellule. Ils sont mesurés avec des couples de détecteurs, sauf pour le dernier point pour lequel on utilise des chambres à fission, les





FIG.5. Répartition radiale des indices de spectre  $\hat{\sigma}_{f~U238}/~\hat{\sigma}_{f~U235}.$ 

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détecteurs ayant un diamètre trop important pour donner une valeur ponctuelle. Les résultats expérimentaux sont en bon accord avec ceux calculés par un code Monte-Carlo [2]:

		Expérience	Calcul
Uranium		$342 \pm 2,5 \cdot 10^{-4}$	$340 \cdot 10^{-4}$
Graphite	Valeur moyenne	$288 \pm 4 \cdot 10^{-4}$	294 · 10 <sup>-4</sup>
	L'imite de la cellule	216 13 . 10	-

Les variations de différents indices de spectre ont aussi été mesurées en trois points de la cellule centrale répartis sur les 10 cm qui correspondent à la hauteur de l'échantillon. On constate une stabilité satisfaisante, compte tenu de la précision, dans la zone de mesure des effets en réactivité:

	<u>- 5 cm</u>	<u>0</u>	<u>+ 5 cm</u>
<sup>238</sup> U/ <sup>235</sup> U	0,998	1,000	0,992
$^{239}$ Pu/ $^{235}$ U	0 <b>, 99</b> 8	1,000	0,996
Th/ $^{235}$ U	1,003	1,000	1,015

# 3.2. Effets en réactivité

Pour comparer les valeurs expérimentales à celles calculées, il est nécessaire de tenir compte de l'hétérogénéité du milieu dans lequel sont effectuées les mesures. Un calcul de cellule en Monte-Carlo [2] nous a permis de comparer les coefficients de réactivité calculés en supposant la cellule composée d'un milieu homogène à ceux obtenus en tenant compte de l'hétérogénéité lorsque la mesure est effectuée dans l'uranium ou dans le graphite. Nous donnons dans le tableau II quelques exemples des résultats obtenus; c'est à l'aide de ces calculs que l'on corrige les valeurs expérimentales pour les comparer à celles calculées en supposant le milieu homogène.

TABLEAU II.	CORRECTI	ONS I	HETERC יו	GENEITE	SUR	LES
EFFETS EN R	EACTIVITE	$\mathbf{PAR}$	GRAMME	RAPPORT	'ES A	
L'URANIUM-2	35					

Corps	Monte-Carlo,	Monte-Carlo, hétérogène			
	homogène	Uranium	Graphite		
Uranium-238	-0,090	-0,088	-0,102		
Carbone	0,229	0,228	0,222		
Fer	- 0, 021	-0,021	-0,020		
Bore naturel	-7,04	-7,38	- 8, 43		
Oxygène	0,112	0,112	0,109		
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Corps	Caractéristiques
Uranium-238	Uranium naturel: $\phi = 12,7$ mm; M=250 g
	Uranium à 20% : $\phi = 12,7$ mm; M = 250 g
	Uranium à 30%: $\phi$ = 12,7 mm; M = 250 g
Plutonium-239	Fils de plutonium à 6% de $^{240}$ Pu; M = 10 g
Thorium	$\phi = 12,7; M = 250 g$
Carbone	Echantillons divers de masse $\leq$ 120 g
Fer	Plaques de 2 mm; M=150 g
Nickel	Plaques de 2 mm; $M = 150 g$
Aluminium	Bloc de $25 \times 25 \times 100 \text{ mm}$ ; M = 150 g
Zirconium	$\phi = 12, 7 \text{ mm}; \text{ M} = 80 \text{ g}$
Bore naturel	Carbure de bore: échantillons de 1 et 2 g
Oxygène	Pastilles d'alumine: $\phi = 12 \text{ mm}$ ; M = 40 g
Sodium	4 réglettes de type MASURCA: M=50 g
Hydrogène	Echantillons d'eau et de polyéthylène d'environ 1 g

# TABLEAU III. CARACTERISTIQUES DES ECHANTILLONS

Les caractéristiques des échantillons sont rassemblées dans le tableau III et l'ensemble des valeurs mesurées et calculées, correspondant à l'effet d'un gramme du corps étudié rapporté à l'effet d'un gramme de <sup>235</sup> U, sont présentées dans le tableau IV.

Pour obtenir les valeurs théoriques présentées, on utilise les constantes d'Hansen et Roach à 16 groupes et le jeu Cadarache à 26 groupes [3] et des calculs à une et deux dimensions en théorie de la diffusion [4] et en théorie du transport [5].

La comparaison des résultats appelle les constatations suivantes:

- La correction d'hétérogénéité par le code Monte-Carlo réduit beaucoup les écarts, surtout lorsqu'elle est importante. Il faudrait dans certains cas effectuer des calculs d'hétérogénéité plus détaillés pour rendre parfaitement compte des conditions de mesure.

- Dans la plupart des cas, c'est le calcul avec le jeu Cadarache qui rend le mieux compte des valeurs expérimentales.

- L'uranium-238, qui correspond à une dilution très faible, n'est pas bien calculé. D'ailleurs, les résultats présentés dans le paragraphe suivant montrent de très fortes variations en fonction du diamètre et de l'enrichissement de l'échantillon, et des calculs plus complets utilisant un code de perturbation hétérogène devraient être effectués pour obtenir une meilleure interprétation de ces variations.

- Le <sup>239</sup>Pu, le carbone et l'oxygène sont en très bon accord ainsi que le thorium si l'on tient compte de la marge d'erreur et des importantes variations que ce corps présente en fonction de la dilution.

Corps	Résultat expe	érimental	Code de t Jeu Hansen	ransport 1 et Roach	Code de diffusion à 1 dimension		
	Brut	Corrigé par Monte-Carlo	2 dimensions 1 dimension		Jeu Hansen et Roach	Jeu Cadarache	
Uranium-238	-0,098 ± 0,002	-0,098	-0,084	-0,079	-0,084	- 0,095	
Plutonium-239	+1,42 ± 0,03	+1,42	+1,40	+1,41	+1,41	+ 1, 44	
Thorium-232	-0,189 ± 0,010	-0,177	-0,166	-0,162	-0,165	-	
Carbone	+0,220 ± 0,002	+0,227	+0,226	+0,232	+0,238	+0,222	
Fer	-0,029 <sup>1</sup> +0,001	-0,029	-0,024	-0,022	-0,023	-0,029	
Nickel	-0,052 ± 0,001	-0,053	-0,136	-0,134	-0,135	-	
Aluminium	-0,005 ± 0,001	-0,005	-0,0010	+0,0031	+0,0025	-0,014	
Zirconium	-0,039 ± 0,001	-0,041	-0,0160	-0,0143	-0,0148	-0,043	
Bore naturel	-8,5±0,2	-7,1	- 6, 66	-6,66	-6,61	-	
Oxygène	+0,104 ± 0,010	+0,107	+0,108	+0,114	+0,117	+0,110	
Sodium	+0,066 ± 0,006	+0,067	+0,043	+0,049	+0,049	+0,043	
Hydrogène	+73,0 ± 2,0	+75,0	+ 53,4	+ 52,6	+ 54, 5	+ 63, 1	

# TABLEAU IV. EFFETS EN REACTIVITE PAR GRAMME RAPPORTES A L'URANIUM-235

- Pour le bore, l'écart est de l'ordre de 6%, mais il faut remarquer que la correction d'hétérogénéité, qui est très importante, ne rendait pas compte de sa répartition exacte dans le graphite de la cellule.

- Les différences trouvées sur le fer et l'aluminium sont certainement dues en majeure partie aux variations de l'importance en fonction de l'énergie, très sensible pour ces corps, et que le calcul ne prend en compte qu'imparfaitement.

- Le nickel et le zirconium donnent des résultats très différents des valeurs calculées, qui ne peuvent être attribuées qu'aux sections efficaces de capture utilisées.

- L'hydrogène et le sodium présentent de gros écarts qu'on peut attribuer aux constantes utilisées pour le calcul des transferts élastiques ou à la mauvaise connaissance de la fonction d'importance, ce qui est plus improbable si on tient compte des bons résultats obtenus avec le carbone et l'oxygène.

## 3.3. Hétérogénéité et effet Doppler

## 3.3.1. Hétérogénéité

Pour étudier les effets en réactivité du <sup>238</sup>U en fonction de la dilution, on utilise des fils d'uranium naturel de 4 mm<sup>2</sup> de section. On a vérifié avec des fils d'uranium enrichi à 93% que, dans la gamme étudiée, l'effet du <sup>235</sup>U était indépendant de la dilution, ce qui confirme les résultats



FIG. 6. Effet en réactivité du <sup>238</sup>U en fonction de la dílution.

théoriques obtenus par Khairallah avec un calcul de perturbation hétérogène [6]. En corrigeant ainsi l'effet du  $^{235}$ U on déduit l'effet du  $^{238}$ U des différentes mesures effectuées avec des barreaux d'uranium naturel de géométries et de densités différentes et des barreaux d'uranium enrichi à 20% et 30%. Les résultats obtenus sont présentés sur la figure 6 et montrent que, dans le cas de l'uranium naturel, l'effet de densité et l'effet de diamètre sont identiques pour une même variation de dilution; par contre, un effet supplémentaire intervient lorsque le  $^{238}$ U est dilué avec le  $^{235}$ U.







FIG. 8. Effet Doppler du <sup>238</sup>U.

#### 3.3.2. Effet Doppler

Les résultats bruts obtenus dans les mesures d'effet Doppler sont représentés sur la figure 7. Les températures indiquées sont une moyenne sur les échantillons, pour tenir compte de la répartition axiale de la température.

Pour l'uranium naturel, métal et oxyde, on effectue une correction de dilatation qui est composée de deux termes: l'un tient compte de la variation de la dilution du <sup>238</sup>U due simultanément à l'augmentation du diamètre et à la diminution de la densité de l'échantillon, elle est obtenue en utilisant la courbe de la figure 6; l'autre tient compte de l'allongement, la répartition axiale des effets en réactivité n'étant pas constante sur la longueur de l'échantillon. Au total, ces deux effets se compensant partiellement, la correction de dilatation est négligeable pour l'oxyde et très faible pour l'uranium naturel métallique (figure 7).

Pour ces deux corps, en négligeant l'effet Doppler du <sup>235</sup>U on a calculé, en utilisant un code de perturbation du premier ordre, la part de la capture du <sup>238</sup>U dans l'effet en réactivité de l'échantillon. Cela a permis d'exprimer l'effet mesuré sous forme d'une variation relative des captures de l'uranium-238 et de comparer les résultats ainsi obtenus avec les résultats des mesures par activation. Les résultats sont présentés sur la figure 8; on constate un bon accord pour les mesures effectuées sur le métal par oscillation et par activation. La différence entre les valeurs obtenues pour le métal et l'oxyde traduit l'écart entre les variations de capture à deux dilutions différentes. Les courbes théoriques tracées sur la même figure sont obtenues à partir d'un jeu de constantes actuellement en cours d'étude [7]. Dans le cas de l'oxyde, on obtient un bon accord avec les mesures, pour le métal on note un écart faible mais significatif entre les deux courbes.

# 3.4. Mesure du rapport des importances

Les deux sources utilisées avaient les caractéristiques suivantes:

Sb-Be :  $2,34 \cdot 10^7$  n/s  $\cdot 4\pi$ Ra-Be :  $8,43 \cdot 10^6$  n/s  $\cdot 4\pi$ .

Les signaux mesurés à différentes puissances, représentés sur la figure 9, sont une fonction linéaire de l'inverse de la puissance, ce qui permet de déterminer la pente de la droite avec une grande précision.

A l'aide du rapport des intensités des deux sources étalonnées par activation d'une solution de sulfate de manganèse, on en déduit que le rapport des importances à ces deux énergies est égal à

$$\frac{I_{Sb}}{I_{Ra}} = 1,067 \pm 0,03$$

La valeur calculée par un code de transport à deux dimensions est de

$$\frac{I_{25 \text{ keV}}}{I_{3 \text{ MeV}}} = 1,085$$

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Le bon accord entre ces deux valeurs nous conduira à développer cette méthode en utilisant comme référence une source de <sup>252</sup>Cf, car la précision du résultat expérimental n'est limitée que par l'étalonnage des sources et la valeur calculée sera plus significative si les spectres d'émission sont mieux connus.



FIG. 9. Oscillations de sources.

# CONCLUSION

Les résultats des indices de spectre comparés à ceux de l'expérience exponentielle HUG montrent que l'on a bien atteint au centre d'ERMINE le spectre caractéristique du réseau.

Les techniques expérimentales utilisées, aussi bien pour les effets en réactivité que pour l'effet Doppler, présentent l'avantage de ne créer aucune perturbation dans le réseau. Les résultats présentés dans ce mémoire montrent que l'on peut ainsi effectuer des mesures précises et interprétables.

1 Les expériences sur le cœur MASURCA de même composition n'étant pas encore effectuées, les coefficients de réactivité n'ont pu être comparés qu'aux valeurs calculées. Cependant, on constate que, si on rend bien compte par le calcul de l'hétérogénéité de la cellule, on obtient un bon accord sur les corps lourds ou très absorbants. Le désaccord qui apparaît sur certains corps ralentisseurs, en particulier l'hydrogène et le sodium, a déjà été constaté dans plusieurs expériences étrangères [8] et se retrouve également pour le cœur MASURCA 1-A [1].

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Nous ne disposons pas encore de suffisamment de résultats pour savoir si ce désaccord est très différent entre le cas de la maquette critique et le cas du cœur à deux zones comme cela a été constaté sur ZPR6 [9]. Cependant, il apparaît actuellement que les difficultés rencontrées dans l'interprétation de certains résultats obtenus au centre d'ERMINE proviennent du fait que le cœur est à deux zones, et non du fait que la zone nourricière est thermique; on pense même que les difficultés seraient les mêmes avec un cœur critique réfléchi.

Ces expériences ont permis de démontrer qu'il était possible d'obtenir des résultats intéressants avec une zone rapide dont le volume représente environ 12% du volume du cœur critique réfléchi. En augmentant légèrement ce volume et en améliorant la zone de transition, on disposera dans le prochain assemblage d'une zone centrale plus importante afin d'étendre les possibilités expérimentales.

On peut ainsi conclure que l'utilisation des assemblages thermiquerapide pour étudier les caractéristiques neutroniques de milieux multiplicateurs rapides peuvent donner les mêmes informations que les cœurs rapides à deux zones, mais ils représentent par contre l'énorme avantage de n'utiliser qu'un faible investissement de matière fissile. Ils constituent ainsi un complément indispensable aux maquettes critiques rapides pour les études de projet de réacteurs surgénérateurs.

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# DISCUSSION

H. SEUFERT: My question concerns the Doppler activation measurements. Did you measure the decay of  $^{239}$ U or  $^{239}$ Np, and how did you discriminate against the background from fast fission?

R. VIDAL: We measured the 74-keV decay gamma of  $^{239}$ U. The detectors placed in the hot and cold samples were irradiated simultaneously by the oscillation method and counted simultaneously; counting started about 15 min after irradiation.

No correction was made for the background; it was not considered necessary since, in the comparison between hot and cold detectors, the fission rate in the two detectors was identical and low compared with the  $^{239}$ U peak. Within the limits of the experimental uncertainty, this finding is supported by the fact that the decay period of the signal corresponds to the half-life of  $^{239}$ U.

E. HELLSTRAND: Your measurements of the importance of neutrons of different energies are very interesting and the accuracy obtained impressive. In connection with the two-dimensional calculations, I should like to know whether the effect of heterogeneity has been taken into account. It should not be negligible, considering the large difference in energy between the Ra-Be neutrons and those from the Sb-Be source.

R. VIDAL: The lattice was only slightly perturbed in this case. The uranium in the central cell was replaced by a graphite channel containing the source. In the interpretation of the results which we have presented, we did not take into account possible differences between the importance of the neutrons in the graphite and that calculated for a homogeneous cell.

W.K. FOELL: Did you, in your analysis of <sup>238</sup>U reactivity and Doppler measurements, allow for the core-sample resonance interaction effects – that is to say, the effects due to differences in absorber density in core and sample?

R. VIDAL: No, we did not. We felt that these effects were negligible for <sup>238</sup>U in the light of the precision achieved in our experiments.

# ANALYSE DES POSSIBILITES D'UNE IRRADIATION MASSIVE DE COMBUSTIBLE POUR REACTEURS RAPIDES DANS BR2, REACTEUR D'ESSAI DE MATERIAUX

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## Abstract - Résumé

ANALYSIS OF THE POSSIBILITY OF LARGE-SCALE IRRADIATION OF FAST REACTOR FUEL IN THE BR2 MATERIALS-TESTING REACTOR. Since September 1965 devices have been used in the BR2 reactor to irradiate fuel pins sodium-cooled under forced circulation, as part of a series of studies on fast reactor fuel designs. The low-energy component of the neutron spectrum incident on these devices is filtered out by boron and cadmium screens. More recently, attempts have been made to design an irradiation device of greater power, which would allow a fast-reactor fuel sub-assembly to be tested to high burn-up rates, under specific spectral and power conditions representative of the next generation of fast reactors. These specifications and the size of the sub-assembly precludes the idea of filtering out the incident spectrum, instead of which this spectrum has to be transformed in a zone at the periphery of the sub-assembly. A device of this type, given the name of SOCRATE, is to be placed in the central experimental cavity (diameter ≈ 200 mm) of the BR2 reactor. It would contain about 380 fuel pins of 6 mm diameter, arranged in a triangular lattice of 7.9 mm pitch and cooled by circulating sodium. The 61 pins at the centre of this lattice form the sub-assembly. At the enrichment adopted, the maximum specific power in the zone containing the sub-assembly is about 1 MW/litre for a maximum linear power of 530 W/cm. A burn-up of 80000 MWd/t could, in principle, be attained within a year. The paper deals in particular with comparative analysis of stresses and damage in the sub-assembly pins, as referred to the properties of the Na-2 reactor core, and concludes that irradiation in SOCRATE could be a significant way of testing a fuel sub-assembly designed for a fast reactor.

ANALYSE DES POSSIBILITES D'UNE IRRADIATION MASSIVE DE COMBUSTIBLE POUR REACTEURS RAPIDES DANS BR2, REACTEUR D'ESSAI DE MATERIAUX. Depuis septembre 1965, des dispositifs d'irradiation de quelques crayons combustibles refroidis par une circulation forcée de sodium sont utilisés dans le réacteur BR2, dans le cadre d'études conceptuelles de combustibles pour réacteurs rapides: la composante de basse énergie du spectre neutronique incident à ces dispositifs est filtrée par des écrans constitués de bore et de cadmium. On a entrepris plus récemment d'analyser la conception d'un dispositif d'irradiation de plus grande capacité, permettant le test d'un sous-assemblage combustible de réacteur rapide jusqu'à des taux d'irradiation élevés, dans des conditions de spectre et de puissance spécifique représentatives des réacteurs rapides de la future génération; ces spécifications ainsi que la taille du sous-assemblage imposent d'abandonner le procédé de filtration du spectre incident au profit de la méthode de conversion dans une zone de couverture disposée à la périphérie du sous-assemblage. Un tel dispositif. dénommé SOCRATE, serait localisé dans la cavité expérimentale (diamètre ≈200 mm) centrale du réacteur BR2; il contiendrait environ 380 crayons combustibles d'un diamètre de 6 mm, disposés suivant un réseau triangulaire d'un pas de 7,9 mm et refroidis par une circulation de sodium; les 61 crayons situés au centre de ce réseau constituent le sous-assemblage. Aux enrichissements adoptés, la puissance spécifique maximale dans la région occupée par le sous-assemblage est d'environ de 1 MW/l, pour une puissance linéique maximale de 530 W/cm; un taux d'irradiation de 80000 MWj/tonne pourrait en principe y être atteint en une année. Le mémoire est particulièrement orienté vers l'analyse comparative, les propriétés du cœur du réacteur Na-2 étant prises comme référence, des contraintes et dégâts d'irradiation dans les crayons du sous-assemblage: cette étude permet de conclure que l'irradiation dans SOCRATE d'un sous-assemblage combustible conçu pour un réacteur rapide serait un test significatif.

# 1. INTRODUCTION

Depuis deux années, des dispositifs d'irradiation de crayons combustibles refroidis par une circulation forcée de sodium sont utilisés dans le réacteur à haut flux BR2 [1]; la majoration de la capacité de ces boucles est en cours d'étude afin de permettre l'irradiation d'un faisceau de sept crayons combustibles. La densité de puissance dans les crayons irradiés dans ces dispositifs peut aisément atteindre et dépasser les niveaux prévus dans les piles rapides : l'enrichissement du combustible et la forme du spectre d'irradiation sont ajustés en conséquence ; la composante à basse énergie de ce spectre peut être plus ou moins éliminée selon que des écrans plus ou moins évolués sont utilisés (B, B + Cd) ; néanmoins les conditions d'irradiation restent très éloignées des conditions rencontrées dans un réacteur rapide et les sollicitations auxquelles est soumis le crayon combustible sont, à cet égard, peu représentatives. L'usage d'écrans absorbants implique en outre une pénalité réactive appréciable pour le réacteur BR2. L'intérêt de l'irradiation dans ces dispositifs ne réside pas dans un test global de la tenue sous irradiation d'aiguilles combustibles destinées à une pile rapide ; en outre, la capacité de ce type de dispositif ne peut que rester modeste.

La demande en volume d'irradiation de combustible pour réacteur rapide allant en croissant, on s'est dès lors attaché à examiner la possibilité d'une irradiation massive dans BR2 d'aiguilles combustibles dans des conditions aussi représentatives que possible. La notion d'irradiation massive a été précisée en prenant pour objectif le test sous irradiation d'un sous-assemblage combustible complet considéré comme une entité mécanique, crayons et structure ; la capacité minimale de cet assemblage a été fixée à 37 crayons combustibles disposés suivant un réseau de type classique pour les centrales rapides futures : plus précisément, des caractéristiques proches de celles du projet allemand Na-2 ont été adoptées. L'usage d'écrans absorbants circonscrits à un tel sous-assemblage, outre qu'il impliquerait une pénalité réactive excessive, ne permettrait pas d'obtenir des conditions d'irradiation satisfaisantes.

Adoptant comme hypothèse première de travail que la géométrie de la matrice du réacteur BR2 ne pouvait être modifiée, c'est-à-dire que les cavités d'irradiation devaient être utilisées telles quelles, on s'est tourné vers l'usage de la cavité centrale du réacteur (diamètre utile d'environ 200 mm) en y disposant un réseau régulier de crayons combustibles refroidis par une circulation de sodium ; la partie centrale de ce réseau accueillerait le sous-assemblage à tester, tandis que la zone périphérique serait constituée de crayons combustibles ayant pour rôle d'assurer, dans la partie centrale du dispositif, des conditions d'irradiation représentatives.

Ce rapport résume l'étude neutronique d'avant-projet du dispositif et est axé sur l'analyse comparée des sollicitations auxquelles serait soumis le sous-assemblage; cet avant-projet neutronique a été développé parallèlement à une étude préliminaire de la conception thermo-hydraulique et mécanique du dispositif; il est loin d'être complet en ce sens que de nombreux aspects conceptuels liés à la neutronique n'ont pas encore été abordés en détail. L'étude paraît néanmoins être arrivée à un stade suffisant pour en dégager les premières conclusions.

Ce dispositif d'irradiation a été baptisé SOCRATE, ce patronyme mettant plus ou moins heureusement en évidence son objectif principal : SOdium Cooled Reactor Assembly Testing Experiment (test d'assemblage pour réacteur refroidi au sodium).

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#### FIG.1. SOCRATE - Vue en coupe du dispositif d'irradiation.

#### 2. DESCRIPTION DU DISPOSITIF D'IRRADIATION

#### 2.1. Vue d'ensemble

Le dispositif d'irradiation (Fig. 1) contient 379 crayons combustibles disposés suivant un réseau triangulaire au pas de 7,9 mm; la zone centrale est occupée par 61 crayons UO2-PuO2 qui peuvent constituer le sous-assemblage combustible à tester quoique, plus probablement, celui-ci ne contiendrait que les 37 crayons centraux, la couronne externe des crayons plutonifères faisant alors partie de la zone de couverture ; dans les calculs effectués jusqu'à présent, la constitution exacte du sous-assemblage n'a pas été précisée et les matériaux de structure correspondants n'ont pas été pris en considération. Parmi les 318 crayons UO2 entourant la zone centrale et constituant la zone de couverture, l'enrichissement est profilé de manière dégressive suivant l'éloignement de l'axe du dispositif ; ce profilage est requis au nivellement de la distribution radiale de la puissance dans le dispositif. Deux solutions ont été retenues : dans la première, l'enrichissement est modifié toutes les deux "couronnes" de crayons : le dispositif contient ainsi au total cinq combustibles différents (régions 1 à 5) ; dans la seconde, l'enrichissement est distinct dans chaque couronne : le dispositif contient donc huit combustibles différents (régions 1 à 8'). Pour ne pas compliquer inutilement l'analyse de l'évolution des caractéristiques neutroniques de la partie utile de la boucle (zone centrale plutonifère) pratiquement insensibles au choix de l'une ou l'autre solution, on a retenu provisoirement la découpe à cinq régions.

Le faisceau des 379 crayons est refroidi par une circulation de sodium pour laquelle deux variantes sont à l'étude : soit une circulation à simple passage, le sodium circulant de haut en bas sur toute la section



FIG.2. Vue en coupe de la zone centrale du réacteur BR2.

occupée par le combustible, soit une circulation à double passage : le sodium introduit à la partie supérieure de la zone de couverture parcourt celle-ci de haut en bas pour refluer ensuite dans la zone centrale ; il est prématuré de faire un choix entre ces deux solutions. Il faut remarquer que les résultats présentés sont cependant plus caractéristiques de la version "simple passage" puisqu'il n'a pas été tenu compte dans le calcul de la paroi de séparation des deux sens de circulation du sodium ni de l'irrégularité que cette paroi introduirait nécessairement dans le réseau combustible.

Le sodium est contenu dans un premier tube d'enceinte séparé d'un second tube constituant l'enveloppe extérieure du dispositif par un matelas de gaz (CO<sub>2</sub>) dont le rôle principal est d'assurer une isolation thermique du tube extérieur ; celui-ci est refroidi extérieurement par la circulation d'eau primaire du réacteur BR2 et est centré dans le chenal expérimental central (diamètre nominal : 203,2 mm) du réacteur. Ce chenal est muni, sur les couvercles supérieur et inférieur de la cuve du réacteur, d'un bouchon d'accès individuel autorisant notamment la solution à simple passage pour la circulation du sodium. La Fig. 2 montre schématiquement la partie intéressante de la matrice en béryllium du réacteur BR2, le dispositif d'irradiation étant en place dans le chenal central ; on a représenté en des positions carac-téristiques quelques-uns des éléments combustibles de BR2 qui constitueraient la configuration de chargement du réacteur, disposée en anneau autour du chenal central ; les 61 crayons plutonifères du dispositif d'irradiation sont représentés par des cercles pleins afin d'illustrer, à l'échelle du réseau de BR2, la taille maximale du sous-assemblage testé.

#### 2.2. Constituants

1. Dimensions du crayon combustible.

Des dimensions hors tout égales ont été adoptées pour la totalité des crayons combustibles.

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: 6,00 mm Diamètre extérieur Epaisseur de la gaine : 0,35 mm Longueur hors tout : 160 cm (couvertures axiales et chambre d'expansion incluses). 2. Matières fissiles. La densité du combustible uniformément distribué dans le volume de la gaine occupé par le combustible (smeared density) a été prise égale à 9,6 soit environ 87 % de la densité théorique. Zone centrale : UO2-PuO2 - environ 15 % en poids de PuO2 dans le mélange - UO2 enrichi à 90 % Composition isotopique (% nombre atomes U + Pu) U-235 : 76,500 Pu-240 : 1,230 **U-238** : 8,500 Pu-239 : 13,641 Pu-241 : 0,123 Pu-242 : 0,006 Zone de couverture : UO2 - enrichissements à optimiser. Le nombre d'enrichissements n'est limité, dans le cadre de l'infrastructure de fabrication considérée, que par la constitution de lots contenant un minimum de 30 crayons de même nature. L'utilisation d'un combustible UO2 classique de même géométrie que les crayons de la zone centrale a été retenue dans cette étude ; cette position pourrait être revue tant au point de vue des dimensions qu'à celui de la nature du combustible (le combustible cermet est notamment pris en considération). 3. Nature du gainage: Acier inoxydable (densité : 7,8 pour T  $\approx 600$  °C) Ni : 13,0 % en poids Composition : Fe : 69,5 % en poids Cr : 16,0Mo : 1,5. 4. Réfrigérant: Sodium (densité : 0,83 pour T = 500 °C). 5. Tubes de force : AISI 304 L, densité : 8,0. ... 6. Isolation thermique : CO2 (30 ata,  $T \approx 180$  °C), densité : 0,036. 3. METHODES DE CALCUL

Deux méthodes de calcul ont été utilisées ; la première est basée sur l'approximation de diffusion, l'autre fait appel à la théorie du transport. Les deux codes correspondants font usage d'une représentation multigroupe des énergies des neutrons, mais avec des découpes différentes ; le modèle géométrique est annulaire et suppose la symétrie de révolution autour de l'axe du dispositif d'irradiation.

3.1. Calculs utilisant la théorie de la diffusion. Le code MODIC II [2] a été associé à une librairie de sections efficaces à 28 groupes d'énergie ; les sections efficaces des 27 premiers groupes sont obtenues par une condensation de la librai-rie GAM [3] ; le groupe thermique couvre les énergies inférieures à 0,683 eV et les sections efficaces correspondantes sont moyennées sur des spectres de Wilkins calculés à partir des paramètres de Westcott et des compositions. La routine de préparation du programme permet de faire intervenir les corrections de structure fine : facteurs de désavantage, atténuation des sections sur la base de l'intégrale de résonance.

A l'actif de ce moyen de calcul, on peut noter :

- le grand nombre d'intervalles géométriques d'intégration
- la facilité de modifications de données : changement d'une composition, ajustement du buckling axial,...

- l'enchaînement automatique des diverses séquences de calcul : préparation des constantes, calculs de criticité, calculs de taux de réaction et d'indices de spectre,...

Au passif, il faut remarquer que le modèle à un seul groupe thermique est peu adapté à l'étude de transitions de spectre aussi importantes que le passage d'un coeur thermique à un réseau rapide.

Dans l'ensemble cependant, les calculs MODIC II permettent de dégrossir l'étude d'un système mixte, tel que SOCRATE dans BR2, en fournissant des résultats suffisamment approchés pour orienter les études plus précises à effectuer en théorie du transport.

#### 3.2. Méthode basée sur la théorie du transport

Le code GMS [4] est un ensemble intégré de routines de calcul pouvant être associé à deux librairies de sections efficaces à 40 groupes d'énergie, dénommées GMS 1 et GMS 2, mises au point à Winfrith. Le code GMS applique la méthode  $DS_n$  de Carlson pour le calcul de criticité ; ce code a été adapté au CEN en FORTRAN IV pour son exploitation sur ordinateur IBM 360-40 ; pour cette étude, l'approximation DS4 a été adoptée et la librairie GMS 2 a été retenue.

La découpe énergétique de la librairie GMS 2 a été fixée dans le cadre d'études de systèmes rapides : les 32 premiers groupes correspondent à la librairie F.D.2 de Winfrith [5] et couvrent les énergies supérieures à 0,683 eV ; leur largeur en léthargie est 0,50 sauf pour le premier groupe ( $\Delta u = 1,00$ ). Mais la gamme des énergies thermiques est détaillée en huit groupes supplémentaires, ce qui permet de représenter fidèlement les spectres à basse énergie et de suivre leur évolution au travers de chaque région ; des transferts entre groupes sont effectués, tant vers les groupes d'énergie supérieure ("upscattering") que vers les groupes d'énergie inférieure ("downscattering") ; le nombre de groupes intervenant dans ce processus dépend de l'isotope considéré.

Le formalisme utilisé pour la préparation des constantes macroscopiques permet l'introduction de facteurs de structure fine de flux dans chaque groupe pour chaque isotope, ainsi que l'usage de sections efficaces microscopiques atténuées pour tenir compte de l'autoprotection des résonances ; le taux d'atténuation est lié à la valeur de la section efficace macroscopique de diffusion potentielle du réseau divisée par la concentration de l'isotope considéré.

Le code dispose également d'une routine de condensation des sections efficaces en un ensemble à nombre de groupes réduit ; la pondération fait appel pour chaque composition à des spectres préexistants, calculés et mis en mémoire, par exemple lors d'un ou plusieurs problèmes précédents ; l'ensemble est complété par des routines fournissant la distribution des taux de fission et divers taux de réaction, ainsi que par une routine d'épuisement.

Le problème majeur de l'étude d'une boucle rapide localisée dans un réacteur thermique est assurément l'analyse de la transition de spectre entre ces deux parties ; on est en droit d'espérer que la découpe de la librairie GMS 2 adoptée pour cette étude puisse permettre un calcul assez précis de cette transition.

Le calcul est effectué en deux étapes :

- calcul de cellule.

On calcule la distribution fine des flux dans une cellule unitaire en milieu radialement infini et on en déduit des facteurs de pondération à appliquer aux constituants pour l'homogénéisation de la région ; ce travail est répété pour chaque région du dispositif. - calcul d'ensemble.

Le calcul de l'ensemble du système SOCRATE-BR2 est ensuite effectué, les différentes régions étant homogénéisées. La carte de



FIG. 3. Distributions radiales de la densité de puissance.

flux obtenue lors de ce calcul GMS est ensuite convertie en distribution de la densité de fission, en spectres normalisés aux différents points et en divers taux de réaction.

#### 4. OPTIMISATION DU CHARGEMENT

Une première étape consiste à optimiser le chargement en combustible du dispositif d'irradiation et d'en calculer les performances à l'état initial ; l'évolution de ces performances avec le taux d'irradiation constituera une seconde étape du calcul.

Cette optimisation vise l'obtention d'une densité de puissance et d'un spectre adéquats dans la zone centrale tout en respectant un critère considéré comme essentiel : les sollicitations maximales doivent rester localisées dans la zone centrale de manière à éviter que l'essai d'irradiation du sous-assemblage ne soit limité par un taux prohibitif de défaillances dans la zone de couverture ; dans les calculs de cet avant-projet, ce critère a été réduit à une expression simple et évidemment insuffisante, imposant que la densité de puissance dans la zone de couverture reste égale ou inférieure à la densité de puissance dans la zone centrale ; l'analyse du critère dans son acceptation complète exige des calculs thermo-hydrauliques couplés à une étude de la structure fine de la distribution de puissance et un examen approfondi des écarts par rapport aux conditions nominales ; cette analyse pourrait conduire, comme cela a été dit au paragraphe 2.2., à modifier la géométrie des crayons, voire la nature du combustible, dans la zone de couverture. La densité de puissance dans la zone centrale est portée près du maximum réalisable:

- en adoptant pour cette zone un combustible à enrichissement très élevé (15 % en poids de PuO<sub>2</sub>, 85 % de UO2 enrichi à 90 % U<sup>5</sup>),
- en poussant les enrichissements du combustible de la zone de couverture aux limites supérieures qu'autorise le critère évoqué cidessus.

En ce qui concerne le combustible de la zone d'irradiation, on reviendra par la suite sur l'influence que peut avoir une forte majoration de l'enrichissement, sur la représentativité de l'expérience d'irradiation.



FIG.4. Répartition énergétique des flux.

Les calculs d'optimisation des enrichissements dans la zone de couverture ont été menés en parallèle pour les deux découpes à cinq et huit régions ; ils ont conduit aux résultats illustrés en traits pleins à la Fig. 3 où on a porté en ordonnée la densité de puissance au niveau du maximum de la répartition axiale, exprimée en kilowatts par litre de mélange, combustible et réfrigérant homogénéisés. Ces niveaux de puissance correspondent à un flux de chaleur maximal de 600 W/cm<sup>2</sup> dans la plaque la plus chaude des éléments combustibles du réacteur BR2. Il a été jugé inutile de poursuivre actuellement ces calculs au-delà de ce stade : on a estimé empiriquement les corrections à apporter aux enrichissements pour obtenir un nivellement plus poussé des maxima de la densité de puissance (courbes en traits interrompus, Fig. 3) : les enrichissements correspondants figurent entre parenthèses ; ces corrections n'affectent pratiquement pas les propriétés de la zone centrale. La suite du calcul a été menée avec la découpe à cinq régions ayant fait l'objet du calcul précis.

La découpe à huit régions permettrait de rendre pratiquement équivalente la densité maximale de puissance dans tous les crayons du dispositif; d'une manière générale, les gradients de puissance dans la zone de couverture s'atténuent quelque peu.

La Fig. 4 permet de comparer les spectres neutroniques au centre du coeur du réacteur Na-2 [6] et au centre et à la périphérie de la zone centrale du dispositif SOCRATE ; les spectres sont représentés sous forme différentielle (flux par intervalle unitaire de léthargie vs énergie ou léthargie) et sous forme intégrale (pourcentage du flux dans



FIG. 5. Répartition énergétique des densités de fission.

le domaine des énergies limité inférieurement par l'énergie E, lue en abscisse). On peut constater, comme en témoignait la distribution de puissance, que le spectre dans le sous-assemblage est fort homogène ; par ailleurs, l'écart entre les spectres Na-2 et SOCRATE s'apprécie mieux en se référant aux courbes représentant le spectre dans une aiguille combustible (85 % en poids de UC enrichi à 90 %, 15 % de PuC ;  $\Phi_{comb}$  : 10,7 mm) irradiée dans BR2 sous écran (B + Cd) et refroidie par une circulation de sodium.

La Fig. 5 est sans doute plus parlante : elle illustre, sous forme différentielle et intégrale, la répartition énergétique de la densité de fission dans les deux milieux Na-2 et SOCRATE. Les deux courbes intégrales sont pratiquement confondues au-dessus d'une énergie de 70 keV : 60 % des fissions dans les deux systèmes sont distribués énergétiquement de la même manière ; elles s'écartent ensuite, la population des fissions induites à basse énergie étant plus élevée dans SOCRATE ; ainsi, la densité de fission en-deçà de 40 eV est nulle dans Na-2 alors que dans SOCRATE, 7,5 % du total des fissions sont produits sous ce seuil énergétique. On a également représenté la courbe intégrale relative au spectre dans l'aiguille irradiée sous écran : l'énergie médiane (50 % des fissions de part et d'autre) est, dans ce cas, égale à 450 eV environ alors qu'elle se situe au voisinage de 180 keV tant dans Na-2 que dans SOCRATE ; on remarque que l'écran (B + Cd) élimine mieux la composante à très basse énergie du spectre BR2 incident que ne le fait la zone de couverture dans SOCRATE. Sur les représentations différentielles, il est intéressant d'analyser les deux "cornes" qui se présentent, dans le domaine des hautes énergies, TABLEAU I. STRUCTURE FINE DE LA DENSITE DE PUISSANCE ET SECTIONS EFFICACES

Paramètre	SOCRATE - zone centrale	Réseau infini de cellules "U naturel"	Réseau infini de cellu- les du type "SOCRATE - zone centrale"		
Creusement, P <u>centre</u> Ppériphérie Distorsion,	0,9944	1,0017	1,0050		
P <u>péri p</u> centre . 100 Pmoyen	+ 0,56 %	- 0,17 %	- 0,50 %		
Sections efficaces moyennes sur énergie	Mo <b>y</b> ennes sur la zone centrale	Moyennes sur la cellule	Moyennes sur la cellule		
$\sigma_{235}$ U, barns	2.096	2.048	1,406		
$\sigma_{239}^{239}$ Pu, id.	2,421	1.893	1,799		
$\sigma^{10}B$ , id.	6,966	2,719	0,892		
$\sigma_{z}^{238}$ U, id.	1,275 10 <sup>-1</sup>	5,372 10-2	1,320 10 <sup>-1</sup>		
Σ <sub>f</sub> , cm <sup>-1</sup> (tous les isotopes)	1,725 10 <sup>-2</sup>	2,857 10 <sup>-3</sup>	1,184 10 <sup>-2</sup>		
Σ <sub>f</sub> , cm <sup>-1</sup> (idem)	SOCRATE - centre 1,609 10 <sup>-2</sup>	Na-2 - centre 2,270 $10^{-3}$			

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d'une manière sensiblement différente d'un milieu à l'autre : cette différence témoigne principalement d'une influence beaucoup plus marquée dans Na-2 de la fission rapide dans l'uranium-238 et du ralentissement inélastique induit par cet isotope.

Ces diverses analyses conduisent en finale au jugement global le plus significatif qui porte sur les gradients radiaux de puissance à l'échelle du sous-assemblage et à celle du crayon combustible.

- Gradient dans le sous-assemblage combustible : La densité de puissance à la périphérie de la zone occupée par un sous-assemblage de 37 crayons combustibles est supérieure de 7 % environ à la densité de puissance en son centre ; ce gradient mesuré dans la région homogénéisée surestime quelque peu la différence de puissance entre le crayon central et un crayon périphérique.
- Gradient dans le crayon combustible : Afin de pouvoir apprécier l'importance relative du gradient de puissance suivant le rayon des crayons combustibles de la zone d'irradiation, on a comparé les trois cas suivants :
  - 1. SOCRATE zone centrale
  - Réseau infini de cellules élémentaires de mêmes dimensions et com-2. position que celles de la zone centrale de SOCRATE, à cela près que l'uranium naturel a été substitué à l'uranium enrichi à 90 % en U-235 ; ce cas a été retenu en vertu de sa similitude avec le coeur du réacteur Na-2
  - 3. Réseau infini de cellules élémentaires identiques à celles de la zone centrale de SOCRATE.

Ces trois cas sont classés dans un ordre de dureté globale croissante

du spectre qui les caractérise. Le tableau I indique, dans les trois cas, les valeurs des creusement et distorsion de puissance dans les crayons combustibles : dans les deux réseaux infinis, on constate une légère majoration de la densité de puissance entre la périphérie et le centre du crayon alors qu'une évolution de sens inverse reste apparente dans SOCRATE ; de toute manière l'écart entre ces valeurs est peu significatif : à même densité de puissance périphérique, les puissances centrales extrêmes ne diffèrent que d'un pourcent.

Les sections efficaces microscopiques citées dans le même tableau per-

- mettent d'apprécier les différences spectrales entre les trois systèmes:  $\sigma_{\rm f} = \sigma_{\rm f} \sigma_{\rm f}^{2350}$  et  $\sigma_{\rm f} = 239$ Pu décroissent de façon monotone quand, globale-ment, la dureté du spectre augmente ;  $\sigma_{\rm a} = 10$ B amplifie fortement les différences spectrales dans le domaine inférieur des énergies épithermiques. -  $\sigma_f 2^{380}$  témoigne d'un spectre sensiblement moins dur à haute éner
  - gie, dans la cellule à uranium naturel.

Les sections macroscopiques de fission illustrent évidemment en premier lieu les différences d'enrichissement ; on peut y voir en outre l'effet de la contamination en neutrons de basse énergie dans le spectre de SO-CRATE, par comparaison avec le spectre d'un réseau infini constitué de cellules identiques ; dans la zone centrale de SOCRATE, la différence entre la section macroscopique moyenne sur la région et la section macroscopique au centre de la même région montre un reliquat de dérive spectrale au travers de la région ; le même effet était également illustré à la Fig. 4 (courbes intégrales).

La comparaison des valeurs de  $\Sigma_f$  dans la cellule-uranium naturel et au centre du réacteur Na-2 ne permet pas de conclusion simple, les deux systèmes restant trop différents : l'enrichissement (Pu) de Na-2 est quelque peu supérieur mais, par contre, ce réacteur contient sensiblement plus de matériaux de structure ; en tous cas, il est permis de considérer que le spectre de Na-2 est moins dur que le spectre du réseau

Seuil énergétique	SOCRATE (n.cm <sup>-2</sup> .s <sup>-1</sup> )	Na-2 (n.cm <sup>-2</sup> .s <sup>-1</sup> )		
3,68 MeV	8,31 10 <sup>12</sup>	1,65 10 <sup>13</sup>		
2,025 MeV	2,95 10 <sup>14</sup>	7,25 10 <sup>14</sup>		
100 keV	1,50 10 <sup>15</sup>	7,86 10 <sup>15</sup>		
2 keV *	1,80 10 <sup>15</sup>	1,30 10 <sup>16</sup>		
460 eV *	1,85 10 <sup>15</sup>	1,34 10 <sup>16</sup>		
100 eV	1,86 10 <sup>15</sup>	1,34 10 <sup>16</sup>		
22 eV *	1,88 10 <sup>15</sup>	1,34 10 <sup>16</sup>		

TABLEAU II. VALEURS ABSOLUES DES NIVEAUX DE FLUX AUX CENTRES DE SOCRATE ET DE Na-2 (NORMALISEES A 500 W/cm DE CRAYON COMBUSTIBLE)

\* La valeur du seuil est approximative en raison des découpes énergétiques différentes utilisées pour le calcul des deux milieux.

infini de cellules du type SOCRATE : ceci permet de conclure que l'écart des creusement et distorsion de puissance dans les crayons du coeur Na-2 et dans les crayons du centre de SOCRATE est inférieur au pourcent, écart maximal apparaissant entre les deux milieux les plus différents au point de vue considéré. Enfin, on peut remarquer, en comparant les valeurs de Ef dans le centre des deux systèmes, Na-2 et SOCRATE, que les "flux totaux" sont, à même densité de puissance, dans le rapport inverse des sections : le flux total est sept fois plus élevé dans Na-2. A cet égard, le tableau II compare les valeurs absolues des flux au-dessus de quelques seuils énergétiques dans Na-2 et SOCRATE, pour une normalisation commune à 500 W/cm de crayon combustible.

SOCRATE. Valeurs caractéristiques.

Caractéristiques initiales, déduites du calcul en DS4 à 40 groupes : (référence : 600 W/cm<sup>2</sup> au point chaud BR2).

- Densité de puissance, zone centrale, au maximum de la distribution axiale : 975 kW/l
- Puissance linéique maximale, crayons zone centrale : 530 W/cm
- Puissance spécifique maximale, idem : 250 W/gr combustible oxyde ou 2.400 W/cm3 de combustible
- Flux calorifique maximal, idem : 280 W/cm<sup>2</sup>
- Puissance totale approximative dans SOCRATE : 10 MW
- Puissance totale BR2 : de l'ordre de 60 MW, suivant chargement pour environ 20 assemblages combustibles
   Apport de réactivité de SOCRATE : de l'ordre de 1 % ∆k/k (état de référence; obturateur Be dans le chenal central).

#### 5. CALCULS D'EVOLUTION

Le code de transport GMS contient une routine de réduction du nombre de groupes ; cette option a été mise à profit pour condenser en 15 groupes les sections efficaces macroscopiques à 40 groupes de chacune des régions de SOCRATE et de BR2, les spectres de condensation étant



FIG. 6. SOCRATE - Spectres neutroniques au centre du dispositif (modèles à 40 et 15 groupes).

les spectres moyens par région du problème initial à 40 groupes. La bonne concordance des deux modèles est illustrée à la Fig. 6.

BR2 fonctionne par cycles de 24 jours, avec un arrêt intermédiaire permettant un remaniement du coeur ; l'intervalle de temps pour la routine d'épuisement a donc été fixé à 12 jours et on a supposé un renouvellement complet du chargement combustible de BR2 tous les 24 jours. Pour les calculs de distribution de flux au début de chaque étape de 12 jours, on impose un laplacien axial rendant le système pratiquement critique ; les flux moyens qui en résultent pour chaque région sont normalisés sur la base du niveau de puissance imposé et sont alors utilisés pour le calcul d'évolution des concentrations isotopiques pendant la durée impartie. Le code répète automatiquement la procédure en prenant comme nouvelles données les teneurs de chacun des isotopes obtenues à la fin de l'étape précédente ; l'hypothèse du renouvellement du coeur BR2 a cependant obligé une fréquente intervention manuelle.

On a représenté à la Fig. 7 les distributions radiales de la densité de fission à différents stades de l'irradiation ; la normalisation de ces distributions est basée sur un flux de chaleur maximum égal à 600 W/cm<sup>2</sup> dans la plaque combustible la plus chaude du chargement BR2. Une réduction progressive du niveau de puissance est observée dans chaque région ; elle est attribuable à la consommation des atomes fissiles et à la réduction des niveaux de flux : ces influences sont atténuées à des degrés divers par une majoration des sections efficaces microscopiques de fission induite par les déformations de spectre.

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FIG. 7. Distributions radiales de la densité de puissance en cours d'irradiation.



FIG. 8. Evolution des sections macroscopiques de fission.

Les régions périphériques du dispositif subissent un appauvrissement relatif important et jouent de moins en moins leur rôle de convertisseur, permettant ainsi à la contamination par les neutrons de bas-



FIG. 9. SOCRATE - Spectres neutroniques au centre. Evolution avec le taux d'irradiation.

se énergie de pénétrer plus profondément dans le dispositif ; cet effet spectral tend à accélérer la vitesse de consommation des noyaux fissiles mais aussi, dans une certaine mesure, le taux de conversion des noyaux fertiles en isotopes fissiles. En outre, la réduction de la densité de fission entraîne une diminution du flux total ; les calculs montrent que les flux totaux des cinq régions du dispositif sont également réduits, en fin d'irradiation, à environ 90 % de leur valeur initiale ; le taux de réduction moins prononcé du niveau de puissance dans la région 3 est donc une conséquence directe d'une réduction relativement moins marquée du  $\Sigma_f$  de cette région.

Les allures différentes de la variation relative des sections efficaces macroscopiques de fission moyennes par région, représentées à la Fig. 8, résultent de la combinaison des influences diverses citées ci-dessus. On observe, par exemple, pour la région 4 une chute accélérée du  $\Sigma_f$  au cours de l'irradiation : relativement aux autres régions, l'augmentation de  $\sigma_f$  compense d'une manière moins marquée la réduction de la concentration en atomes fissiles. Les calculs montrent par ailleurs que l'accumulation de <sup>239</sup>Pu dans la région 3 est plus marquée que dans les autres régions de la zone de couverture.

La zone de couverture offre dans son ensemble une barrière encore bien suffisante pour que l'accroissement de la contamination de la zone centrale par les neutrons de basse énergie soit indiscernable ; ce phénomène est illustré à la Fig.9 où les spectres sur l'axe du dispositif ont été représentés à l'état initial et en fin d'irradiation; on constate également à la Fig.7 que le profil radial de la distribution de la densi-



FIG. 10. SOCRATE - Spectres neutroniques au centre, à l'échelle des densités de puissance.

té de puissance n'y est pas sensiblement modifié. La Fig.lO met en évidence la gamme d'énergie dans laquelle la réduction du flux total ( $\sim 10$  %) au cours de l'irradiation est la plus marquée ; les deux histogrammes de cette figure ont été normalisés proportionnellement aux densités de puissance en début et en fin d'irradiation ; on constate que la partie à basse énergie est pratiquement non affectée.

Au cours de 240 jours effectifs de fonctionnement (c'est-à-dire en pratique une année), on pourrait en principe atteindre dans la zone centrale un taux d'irradiation de l'ordre de 80.500 MWj/tonne <sup>1</sup> au niveau du maximum axial. L'évolution temporelle des taux d'irradiation dans chacune des régions et des pourcentages d'atomes lourds consommés est détaillée dans le tableau III. La Fig. 11 montre l'évolution en fonction du temps des concentrations des divers isotopes du plutonium et de l'uranium dans la zone d'irradiation.

Il est important de souligner également que la dérive de réactivité associée à l'évolution du dispositif pendant un an n'est que de 1,5 10-2  $\Delta k/k$  environ.

#### 6. DEGATS D'IRRADIATION.

Si les contraintes dans le crayon combustible peuvent être considérées comme équivalentes dans les deux systèmes comparés, SOCRA-TE - zone centrale et Na-2 - coeur, il faut examiner s'il en est de même quant à la résistance que peut leur opposer le gainage au cours de l'irradiation.

 Les taux d'irradiation sont rapportés à la tonne de combustible (U + Pu) métal.

#### SM-101/24

Parmi les deux mécanismes de fragilisation du matériau de gainage, déplacements atomiques et formation d'atomes de gaz, il est bien admis que le premier a un effet résultant négligeable aux températures voisines de 600 °C atteintes dans les gaines des milieux considérés. En ce qui concerne la fragilisation induite par l'accumulation de gaz aux joints des grains, on considère que l'hydrogène, produit dans les réactions (n,p), a une influence secondaire vis-à-vis de l'hélium produit par réaction  $(n,\alpha)$ ; l'hydrogène formé diffuse en effet plus facilement hors du matériau de gainage.

Dans l'acier de gainage adopté ( $\overline{F}e: 69,5$ ; Cr: 16,0; Ni: 13,0; Mo: 1,5 % en poids) on s'est donc attaché à examiner les mécanismes de production d'hélium afin de pouvoir en comparer principalement les taux initiaux de production dans les deux milieux.

Trois sources d'hélium ont été traitées séparément :

- réaction  $(n, \alpha)$  avec les constituants de l'acier : Fe, Ni et Cr.
- réaction (n,α) avec l'azote (0,06 % en poids) inclus dans l'acier.
  réaction (n,α) avec le bore présent dans l'acier à l'état d'impureté et apporté principalement à l'alliage par le nickel (50 ppm de bore dans le nickel, soit 6,5 ppm de bore dans l'acier).

La variation énergétique des sections efficaces  $\sigma(\mathbf{E})$  des deux premières réactions a fait l'objet d'une analyse bibliographique : seule la partie à pseudo-seuil de la section efficace dans le domaine des énergies neutroniques les plus élevées vaut d'être prise en considération dans les milieux envisagés.

Pour chaque réaction, un seuil effectif,  $E_{eff}$ , et la section efficace effective correspondante,  $\sigma_{eff}$ , sont déterminés ; cette section a une valeur constante au-dessus du seuil et nulle en-deçà.

On recherche, suivant une technique dérivée de la méthode de Grundl et Usner [7], le couple de valeurs ( $E_{eff}$ ,  $\sigma_{eff}$ ) qui, tout en satisfaisant l'expression

$$\nabla_{\text{eff}} \int_{E}^{\infty} \sinh \nabla \sqrt{E} \cdot e^{-\beta E} \cdot dE = \int_{O}^{\infty} \nabla(E) \cdot \sinh \nabla \sqrt{E} \cdot e^{-\beta E} \cdot dE$$

dans laquelle

 $v = \sqrt{2}$  et  $\beta = 1$  (spectre de Watt)

est à la fois le plus insensible à des variations du paramètre y et du paramètre  $\beta$ , c'est-à-dire à une distorsion, par rapport au spectre de fission de l'uranium-235, du spectre dans lequel le taux de réaction doit être calculé. On détermine ainsi les valeurs suivantes : Acier - Eeff : 6,85 MeV ;  $\sigma_{eff}$  : 26,5 mb

Azote	-	Eeff	:	2,025 MeV	;	<sup>o</sup> eff	:	223 mb	

Les spectres neutroniques ci-après ont été adoptés : - SOCRATE : axe de la zone centrale homogénéisée (GMS, 40 groupes) - Na-2 [6] : axe du coeur de la pile homogénéisé (NUSYS, 26 groupes). Ces spectres ont été normalisés à l fission/cm<sup>3</sup>.s puisque aussi bien les deux systèmes peuvent être considérés comme également capables d'une même puissance spécifique. Les neutrons contenus dans le groupe énergétique où venait s'intercaler le seuil effectif de la réaction  $(n, \alpha)$ considérée ont été supposés répartis, entre les limites énergétiques de ce groupe seulement, suivant un spectre de fission <sup>235</sup>U. Les précisions avec lesquelles les sections différentielles  $(n, \alpha)$  sont connues (de ± 20 à 50 % suivant l'isotope) autorisent à considérer ces diverses hypothèses comme satisfaisantes.

Région	Durée d'irra- diation(jours effectifs)	24	48	72	96	120	144	168	192	216	240
1	MWj/tonne	8.690	17.220	25.630	33.860	41.930	49.910	57.725	65.415	73.000	80.465
-	$\frac{\frac{N_{o} - N(t)}{N_{o}}(\%)}{N_{o}}$	1,17	2,32	3,46	4,57	5,66	6,73	7,79	8,83	9,85	10,86
2	M∀j/tonne	8.865	17.990	26.600	35.060	43.400	51.630	59.770	67.830	75.830	83.750
	$\frac{\frac{N_{o} - N(t)}{N_{o}}(\%)}{\frac{N_{o}}{N_{o}}}$	1,26	2,50	3,73	4,93	6,11	7,27	8,42	9,56	10,69	11,80
	M∀j/tonne	7•350	14.610	22.555	29.655	36.685	43.660	50.595	57.490	64.345	71.160
_	$\frac{\frac{N_{o} - N(t)}{N_{o}}}{N_{o}}$	1,03	2,04	3,03	4,02	4,98	5,94	6,88	7,83	8,75	9,67
4	MWj/tonne	7.240	14.285	21.145	27.810	34.280	40.550	46.630	52.500	58.160	63.600
4	$\frac{N_{o} - N(t)}{N_{o}}$ (%)	0,98	1,93	2,84	3,72	4,57	5,39	6,18	6,94	7,66	8,36
5	MWj/tonne	7.500	14.500	21.015	27.065	32,690	37.905	42.745	47.250	51.450	55.390
2	$\frac{\frac{N_{o} - N(t)}{N_{o}}(\%)}{\frac{N_{o}}{N_{o}}}$	1,00	1,90	2,73	3,50	4,18	4,82	5,42	5,96	6,53	6,95
*	Plus précisé	ment : 2	35 <sub>0</sub> 238 <sub>0</sub>	239 <sub>Pu</sub>	240 24 Pu 24	+1 Pu et 2	242 Pu				

TABLEAU III. TAUX D'IRRADIATION MOYENS PAR REGION ET POURCENTAGES DU NOMBRE D'ATOMES (U+Pu)\* PERDUS PAR FISSION ET CAPTURE

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FIG. 11. Concentrations isotopiques dans la zone centrale.

TABLEAU IV.	TAUX DE REACTION $(n, \alpha)$ DANS SOCRATI	E
ET Na-2		

Constituant	Taux de réaction po en 10 <sup>-6</sup> /0	SOCRATE Na-2 . 100	
	SOCRATE	Na-2	en %
Acier	69,3	137,3	50,5
Azote	49,9	122,7	40,7
Bore	21,6	75,9	28,4
Total	140,8	335,9	41,9

(\*) Par cm<sup>3</sup> du milieu homogénéisé : seul l'acier du gainage est pris en compte.

Le taux de réaction dans le bore,  $\sum_{i=1}^{n} (\Sigma_{a} \Phi)_{i}$ , a été calculé en utilisant les bibliothèques du bore-lO associées aux codes GMS et NUSYS.

Les taux de réaction initiaux dans l'acier, l'azote et le bore sont donnés au tableau IV : le taux global de réactions  $(n,\alpha)$  dans SOCRATE, en début d'irradiation, représente 42 % du même taux de réaction dans le coeur du réacteur Na-2.

Ne disposant pas de données précises quant à l'évolution dans Na-2 des spectres et niveaux de flux en fonction du taux d'irradiation, on n'a pu que vérifier assez grossièrement que ce pourcentage n'était pas sensiblement modifié en fonction du taux d'irradiation : un effet de compensation entre tendances opposées est d'ailleurs observé. Ce pourcentage peut dès lors également représenter le rapport entre les taux intégrés de réaction  $(n,\alpha)$ , à un taux d'irradiation donné (pour un taux d'irradiation de 50.000 MWj/tonne, environ 1017 réactions  $(n,\alpha)$ par cm<sup>3</sup> du milieu homogénéisé).

Considérant les incertitudes inhérentes à cette analyse, on ne lui attribuera que le mérite de situer un ordre de grandeur : la production d'hélium dans le gainage des crayons combustibles est le double environ dans le coeur Na-2 de ce qu'elle est dans la zone centrale du dispositif SOCRATE.

Dans l'état actuel des connaissances sur les mécanismes de fragilisation, cette différence ne paraît pas significative : on peut donc conclure que les différences des niveaux de flux entre les deux systèmes, mises globalement en évidence par la majoration d'enrichissement requise pour obtenir dans SOCRATE une densité de puissance équivalente à celle du coeur Na-2, ne pénalise guère l'expérience d'irradiation en ce qui concerne la fragilisation du gainage.

Il ne manque d'ailleurs pas d'auteurs qui considèrent que les phénomènes de fragilisation envisagés ici sont, dans une très large mesure, uniquement dépendants de la densité de fission.

#### 7. AUGMENTATION DES PERFORMANCES

En parallèle avec les études d'optimisation et d'évolution du dispositif SOCRATE, on a examiné les moyens d'en augmenter les performances, soit par transformations dans le réacteur BR2, soit par modifications de la conception de la zone de couverture. Cette analyse conduit à ne retenir en pratique qu'une solution qui soit peu coûteuse et réalisable sans aucun développement technologique particulier : elle conduit à majorer le niveau de flux incident à la périphérie du dispositif d'irradiation et, par là, la densité de puissance dans le dispositif.

Le niveau de flux incident sera plus élevé, pour un même flux calorifique maximal au point chaud du chargement de BR2, si l'enrichissement ou la teneur en combustible dans les plaques des éléments combustibles est réduit : ces deux options conduisent d'ailleurs à des effets très semblables dans le domaine de variation acceptable.

Ce domaine est en effet limité par l'obligation de charger dans le BR2 un plus grand nombre d'éléments combustibles pour atteindre un même investissement de réactivité : la puissance totale du BR2 en est accrue et le plafond de la capacité (80 MW) des échangeurs primaires est rapidement atteint.

Compte tenu de cette limitation, une réduction à 70 % <sup>235</sup>U de l'enrichissement actuel (90 % <sup>235</sup>U) du combustible de BR2 permettrait d'accroître de 15 % environ la densité de puissance dans la zone d'irradiation, sans y modifier sensiblement le spectre neutronique ; la puissance linéique maximale dans les crayons du sous-assemblage central de SOCRATE atteindrait dès lors 600 W/cm environ.

On a par ailleurs vérifié que la réduction d'enrichissement dans le coeur BR2 conduit à une modification acceptable de l'effet réactif associé à une réduction de la densité du réfrigérant (coefficient de vide).

#### 8. DISCUSSION

En ce qui concerne les divers aspects sous lesquels le caractère représentatif de l'expérience d'irradiation a été analysé, il reste à examiner les conséquences de la majoration de l'enrichissement du combustible sur le spectre des produits de fission : la dissemblance de ces SM-101/24

spectres requiert un examen comparatif des situations résultant des affinités chimiques différentes des produits formés vis-à-vis de l'oxygène. Il faut d'ailleurs remarquer que ce problème n'est pas particulier aux irradiations dans SOCRATE : un artifice semblable est également utilisé pour les irradiations dans des réacteurs rapides de combustible destiné aux grands projets actuels.

A l'échelle du crayon combustible, la distribution radiale quasi uniforme de la densité de puissance assure une répartition des températures et des produits de fission similaire à celle qui caractérise les réseaux purement rapides : à ce point de vue, l'évolution métallurgique du combustible dans SOCRATE sera donc représentative.

En ce qui concerne le sous-assemblage combustible, le gradient transversal de puissance est modéré ; les études bidimensionnelles en cours permettront de préciser les distributions axiales de puissance : à cet égard, le déplacement cyclique des barres de contrôle du réacteur BR2 tendra à uniformiser la puissance intégrée sur une tranche appréciable de la hauteur des crayons.

Les conditions de travail des crayons situés dans la zone de couverture doivent être investiguées en détail : à cet égard, la structure fine de la densité de puissance dans la section droite de ces crayons est une donnée fondamentale ; des techniques éprouvées [1] appliquées à un modèle du dispositif localisé dans BRO2 permettraient de déterminer cette caractéristique avec la précision requise. Plus généralement d'ailleurs, la disponibilité de BRO2, modèle nucléaire précis du réacteur BR2, est considérée comme un atout essentiel à la réalisation du dispositif d'irradiation : la plupart de ses caractéristiques peuvent y être confirmées.

Sur le plan de l'analyse de la sécurité, les études ont été entamées ; on a ainsi vérifié que SOCRATE localisé au centre de la matrice du réacteur BR2 forme un système très largement sous-critique en l'absence d'éléments combustibles du BR2 ; l'immersion dans l'eau légère du faisceau des 379 crayons combustibles conduit à un facteur de multiplication de l'ordre de 0,80 : cette situation est évidemment due à la forte sous-modération de ce système et à sa géométrie défavorable ; d'autre part, le déchargement du dispositif accompagné d'une introduction massive d'eau dans la cavité centrale du réacteur conduit à un déficit réactif important.

D'une manière plus générale, la cinétique du système couplé, SOCRATE-BR2, est dominée par les caractéristiques du BR2 ; l'apport réactif du dispositif est en effet très faible.

Enfin, l'étude des situations anormales dans la partie rapide du système, telles que la réduction de densité ou la vidange du sodium,est à entreprendre ; il en est de même des effets réactifs associés à la fusion du combustible.

Par ailleurs, la technologie particulière des circuits sodium dans le réacteur BR2 a été éprouvée à l'occasion de la réalisation de plusieurs dispositifs antérieurs [1]; la présence d'une quantité de sodium sensiblement supérieure dans le système primaire et dans la piscine du réacteur BR2, inhérente à l'évacuation d'une puissance élevée, situe évidemment la complexité du problème à un niveau très différent.

#### 9. CONCLUSION

Cette étude avait pour but d'analyser les conditions d'irradiation dans SOCRATE afin de pouvoir apprécier si l'irradiation d'un sous-assemblage combustible conçu pour un réacteur rapide serait un test significatif : cette étude apporte une réponse affirmative qui justifie de poursuivre l'analyse des caractéristiques du dispositif afin de pouvoir juger s'il est réalisable dans un délai de deux ans et dans des conditions financières acceptables. L'étude des contraintes thermiques dans les crayons combustibles localisés dans les importants gradients de puissance de la zone de couverture est très probablement l'élément d'appréciation le plus critique.

Tant pour ce problème particulier que, d'une manière générale, pour l'ensemble du projet, la réalisation d'un modèle nucléaire du dispositif dans BRO2 paraît être un passage obligé.

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# FAST-THERMAL COUPLED SYSTEM FOR INTEGRAL MEASUREMENTS OF FISSION-PRODUCT CROSS-SECTIONS\*

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#### Abstract

FAST-THERMAL COUPLED SYSTEM FOR INTEGRAL MEASUREMENTS OF FISSION-PRODUCT CROSS-SECTIONS. In the framework of the co-operation of the West Germans, Belgians and Dutch on fast-breeder research a feasibility study has been initiated on the possibilities for integral measurements of fission-product cross-sections for fast reactor systems. A recent study (Rep. GEAP-4472) suggests that uncertainties may exist regarding, for instance, coolant void coefficients of 10\$ for a sodium-cooled and of 20\$ for a steam-cooled design. Some of these uncertainties are caused by inaccuracies in cross-sections for fissile and fertile isotopes, and can be reduced considerably by results from critical experiments. The remainder (up to 4\$ for sodium and up to 6\$ for steam-cooled breeders) is mainly due to inaccuracies in cross-sections of fission products.

Because of the high radioactivity, direct measurements in a critical experiment with actual fission products are impossible. The use of mock-ups of fission products still leaves big uncertainties because of the differences in isotopic compositions of natural elements occurring in fission products. It is therefore proposed to convert the existing, heavily shielded, light-water critical experiment KRITO at the Petten site of the Reactor Centrum Nederland into a coupled fast-thermal system, in the fast zone of which by oscillator methods integral cross-sections of actual fission product mixtures, elements and nuclides, are to be measured by oscillator methods. To specify the compositions of the fast zone a number of 26 group diffusion calculations have been performed based on the ABBN set.

Regarding the influence of fission products on the coolant void effect, which is the major effect to be looked at, it appeared that 90% of the influence is caused by neutron absorption in the range of 2 keV to 50 eV for a sodium-cooled 1000-MW(e) breeder and in the range of 6 keV to 10 eV for a steam-cooled 1000-MW(e) breeder. Compositions were therefore sought for the fast zone giving about the same reactivity versus energy response to a sample of fission products introduced in the centre of the fast zone. It was found that compositions consisting of 90% enriched uranium with graphite in atomic ratios varying from 25 to 50 graphite atoms per <sup>236</sup>U atom, and with a slight addition of polyethylene (from zero to one H atom per 235U atom) will produce the wanted spectra and energy-reactivity response for the oscillator. For such compositions the sensitivity of the oscillator will be of the order 1 to  $3 \times 10^{-6} \delta k/k$ per gram of fission product mixtures for a coupled system which derives 50 to 75% of its reactivity from the fast zone. A design has been made which can accomodate the variable fast zone sizes needed for the various spectra. The programme of the measurements is outlined. At the same time a small heavily shielded facility is being designed for chemical separation of fission product elements from reactor wastes and from <sup>239</sup>Pu and <sup>235</sup>U irradiations in the high flux reactor at Petten. In addition to the fission product cross-sections, integral cross-sections for other materials and even Doppler coefficients could be measured in the proposed system.

# 1. INTRODUCTION

In the framework of the co-operation of the West Germans, Belgians and Dutch on fast-breeder reactor research, a study has been undertaken by the RCN on the feasibility of integral measurements of the crosssections of actual fission products in neutron spectra of interest for fast reactors.

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Fission products constitute one of the sources of inaccuracies in the prediction of fast reactor behaviour, A recent study [1] suggests that, in general, uncertainties may exist, for example, in coolant void coefficients of 10 \$ for a sodium-cooled fast reactor and of 20 \$ for steam cooling. Part of these uncertainties can be attributed to inaccuracies in the cross-sections of the fissile and fertile isotopes. The uncertainties due to the inaccuracies in the knowledge of the fission product cross-sections may range up to 4 \$ or 6 \$ for a sodium or a steam-cooled breeder, respectively.

It is to be expected that the inaccuracies in the cross-sections of fertile and fissile materials will be reduced considerably in the coming years as a result of the differential cross-section measuring programmes under way at several laboratories and by the results of critical experiments.

The same is true for other core materials. The situation looks less promising for fission products. For a number of more important fission product isotopes most or even all experimental information is lacking. For this reason one has to resort to theoretical estimates (see, for example, Ref.[2]). As the number of important fission product isotopes, daughter products and capture products amounts to about 90 it is unrealistic to expect results of differential cross-section measurements for all these isotopes in the foreseeable future. Moreover, the high activity of a number of these isotopes hampers direct measurements. Also, a number of these isotopes are available only rarely or not at all in enriched form.

Therefore integral measurements on actual fission products could contribute to the reduction of existing uncertainties. For the measurements of, for example, the effect of fission products on coolant void coefficients, the use of actual fission products in a critical experiment is very difficult because of the radioactivity. For this purpose mock-up fission products have been devised. The most general and elaborate mock-up system is described in Ref. [3]. In this mock-up only 60% of the actual fission products (with half-lives longer than one year) can be represented correctly; the remainder have to be simulated by naturally occurring and not too expensive materials. As this simulation is based on the previously mentioned theoretical estimates of cross-sections a rather large uncertainty is still present.

A further experimental study on the reactivity effects of actual fission products seems therefore necessary. Because of the high activity (1 kCi per gram fission products of a few months old) and of the scarcity of separated isotopes only the use of small samples is feasible. This leads directly to a central reactivity value measurement by, for example, pile oscillator techniques in a reactor system in which flux and adjoint flux spectra have a sufficient correspondence with the spectra in an actual reactor.

The study made by the RCN on how such measurements could be performed at Petten has led to a design-proposal for a fast-thermal coupled facility replacing the present thermal critical facility KRITO [4], located in a heavily shielded bay at the RCN site at Petten. In the central fast zone of this coupled facility the measurements on bulk fission product samples, fission-product elements and fission-product isotopes will be performed by a pile oscillator technique. Some considerations leading to this design, some results from the supporting

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calculations, and the proposed experimental programme are briefly discussed in the following pages.

# 2. EFFECTS OF FISSION PRODUCTS IN POWER BREEDER REACTORS

Two types of effects of fission products on the behaviour of a power breeder reactor have to be considered.

# 2.1. Direct effects

The first type is direct effects, e.g. on reactivity and breeding ratio. Here the effects of fission products are directly related to the ratio of the reaction rates (mainly absorption) of the fission products to the reaction rates in the core constituents (especially fuel) at the same energy. In this case the energy range of interest for the knowledge of the fission product cross-sections is that range in which most of the fissioning and breeding takes place.

# 2.2. Difference effects

The second type is difference effects, i.e. the effects of fission products on coolant void reactivity coefficients. In this case the differences in the reaction rates of fission products in the non-voided and voided reactor are important. Here a sort of "difference spectrum" is thus the quantity of interest.

As the difference effects were thought to be more important, and as the requirements for measurements concerned with these effects are probably more difficult to meet, they received prime attention.

To obtain some reference data for the magnitude and energy dependence of the fission product influence on void coefficients, a few calculations were made for the reference designs NaI [5] and DI [6]. All calculations referred to in this paper were done in 26 groups with one-dimensional diffusion programmes EDDI and FREDDI written in ALGOL 60 for the Electrologica X8 computer of the RCN. All cross-sections used were taken without modifications from the 26 group library of ABBN [7].

By one-dimensional first-order perturbation calculations the over-all reactivity effects of adding fission products to the non-voided and to the 100% voided core of the NaI reactor has been determined. From this calculation estimates were obtained for the effect of fission products on the void coefficient for each energy and each perturbing phenomena (absorption, scattering, etc.), separately. In this perturbation calculation  $10^{20}$  pairs of fission products per cm<sup>3</sup> were homogeneously added to the core. This corresponds to a homogeneous burn-up of about 12 000 MWd/t. The reactivity loss due to the addition of these fission products to the non-voided core was - 0.90%, and addition to the 100% voided core produces a loss of only - 0.56%. The difference, + 0.34%, can be considered as a measure of the positive contribution of fission products to the void coefficients.

Inspection of the reactivity contributions per energy group as given in the Figs 1 and 2 reveals that nearly 90% of the fission product effect is caused by absorption in the energy region 2 keV to 50 eV. The other



FIG.1. Contribution of each energy group to the reactivity loss due to absorption of fission products added to the normal and voided core of the NaI reactor.



FIG.2. Breakdown of the contribution of each energy group to the effect of fission products on the void coefficient for the NaI reactor.

contributions comprise leakage and transport effects in the region 2 MeV to 0.2 MeV, contributing about +10%; absorption about 10 keV, contributing about +10%; and absorption in the sodium resonance at 2.8 keV, contributing about -10%.

For the steam-cooled reactor DI analogous results are given in Fig. 3. From this it is clear that here most of the effect is due to absorption in the energy region 6 keV to 5 eV.



FIG. 3. Contribution of each energy group to the reactivity effects due to fission products in the DI reactor.

From these figures, however qualitative they may be, it is clear that for the measurements of absorption cross-sections of fission products most emphasis should be placed on the range from a few keV down to several eV, i.e. the resonance region. It is in just this region where data for most of the fission product cross-sections are lacking, and where reliable predictions based on nuclear theories arehard to make.

## 3. COMPOSITION STUDIES

In a survey study, using first-order perturbation calculations a large number of compositions of fast systems have been tested on their response of reactivity versus energy to a small sample of mixed fission products introduced in their centre, in order to find compositions that might produce responses as given in the Figs 2 and 3. A high "sensitivity" ( $\delta \rho$  per gram of fission products introduced in the centre) is wanted

# TABLE I. SURVEY OF SEVERAL COMPOSITIONS AND THEIR CHARACTERISTICS

1	2	3	4	5	6	7	8	9	10	11
		Appro:	ximate							
		VOI. % OI		r	Ddilut		mass			
Composition	CH <sub>2</sub>	С	11 005	D 10	<sup>n</sup> II-235	R <sub>c</sub>	U-235	δρ/g	ΔE	Etop
number	ρ=1	$\rho = 1, 5$	0-235	B-10	0 200	(em)	(kg)	X 10		<i>F</i>
1	36	-	64	-	1:1	12.3	90	- 42	400 keV-4eV	4-0,01 keV
2	53	-	47	-	2:1	12.8	77	- 195	4 keV-2eV	1-0.01 keV
3	63	-	37	-	3:1	14	80	- 240	1 keV-1eV	1-0.01 keV
4	74	- 1	26	-	5:1	15.3	76	- 340	200 - 2eV	50 <b>-</b> 4 eV
4, 1	74	-	26	16.7	5:1	18	124	- 125	Many peaks	
4, 3	74	-	26	50	5:1	25	332	- 32	Many peaks	
5	80	- 1	20	-	7:1	15.5	60	- 480	Many peaks	
6	85	-	15	-	10:1	15.7	46.5	- 597	Many peaks	
6, 1	85	-	15	16.7	10:1	29	284	- 60	Many peaks	
7	94.4	-	5.6	-	30:1	14.8	14.4	<del>-</del> 1400	Many peaks	
7, 0, 1	94.4	-	5.6	1.7	30:1	22.5	51	- 354	Many peaks	
7,0.3	94.4	-	5.6	5	30:1	31	134	- 115	Many peaks	
8	98.2	-	1.77	-	100:1	15.7	5,1	- 1730	Many peaks	
8,0.1	98.2	-	1.77	1.7	100:1	35	56	- 151	Many peaks	
9	-	86.5	13.5	· -	10:1	34	416	- 3.7	1 MeV-2 keV	100 <b>-</b> 2 keV
10	-	92.7	7.3	-	20:1	42.4	432	- 13	100 <b>-</b> 0,2 keV	10-2 keV
11	-	95	5	-	30:1	47.7	430	- 20.3	50keV-50 eV	5-0.2 keV
12	-	97	3	-	50:1	57	440	- 30.3	10-0.01 keV	0.2 keV
12, 0.1	-	97	3	1.7	50;1	59	490	- 22.9	10-0.02 keV	0.2 keV
12, 0.3	-	97	3 <sup>.</sup>	5	50:1	70	820	- 11.2	10 <b>-</b> 0. 05 keV	0.2 keV

 $^{a}\,$  B-10 is given in mg elementary B-10 per cm .

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because, due to the high radioactivity of actual fission products, the amount of sample that can be handled will always be small. Therefore, in the first place most of the reactivity in the measuring system should be derived from the energy region where most of the absorption in the fission products takes place; secondly, the system should contain no or only a limited amount of materials with high resonance capture, such as, for example,  $^{238}$  U.

This leads to compositions with graphite or hydrogen (or combinations thereof) as slowing-down materials and with highly enriched uranium as fuel.

To cut off a too high epithermal contribution a small amount of boron could be added.

Some results of the calculations are given in Table I, where a number of material compositions are given together with some pertinent data. Columns 7 and 8 give the radius and mass of the bare spherical system with the given composition. Column 9 shows the "sensitivity" of the composition in terms of the central reactivity value due to the introduction of 1 g of a fission product mixture to the bare sphere. Columns 10 and 11 show the energy range of the reactivity response and the location of the top of this response. Some of the responses are also given in the Figs 4 and 5.

The effect of adding some hydrogen in the form of polyethylene has also been studied. Results are given in Figs 6 and 7 and Table II.

On the basis of these results the compositions 1:30 C, 1:50 C and 1:75 C + 1 H have been selected as reference compositions for the design. The spectra produced by these compositions are given in Fig. 8 together with central spectra of the NaI and DI reactors. From this it is clear that the measurements have to be performed in several spectra, i.e. several compositions will have to be employed.

In all these calculations all materials were supposed to be homogeneously mixed, so the results given here have to be corrected for the inevitable heterogeneity that in fact occurs. It appeared from separate calculations that the heterogeneity does not essentially alter the picture given above. However, considerable attention should be paid to selfshielding and mutual shielding effects in and around the sample.

## 4. COUPLED FAST-THERMAL SYSTEM DESIGN

The facility to be built by the RCN to measure fission product crosssections will be a fast-thermal coupled system. The arguments for this are: firstly, lower amounts of enriched fuel are needed for a coupled system compared with an all-fast system; secondly, the longer mean neutron life-times in a coupled system eases the safety problems; and thirdly, this facility will have to replace an existing light-water critical experiment. The feasibility and safety of a coupled fast-thermal system have been clearly demonstrated by the STARK facility [8].

To have the full advantages of the safety characteristics of water reactors, the thermal zone of our facility will be water-moderated. For low thermal fuel inventory and high fast flux-to-power ratio, highly enriched plate-type fuel will be used for the thermal zone, which is a cylindrical annulus surrounding the fast zone. The thickness of this



FIG.4. Reactivity response versus energy due to fission products introduced in compositions 1: 5 H.



FIG. 5. Reactivity response versus energy due to fission products introduced in compositions 1 : 50 C.



FIG.6. Reactivity response versus energy due to fission products introduced in compositions 1: 30 C+H.



FIG.7. Reactivity response versus energy due to fission products introduced in compositions 1:75 C + H.

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Composition		Atom rati	os		Critical radius	Critical mass	δρ/g scatt.	δρ/g abs.	δρ/g total
number	U <b>-</b> 235	U-238	с	н	(cm)	(kg U-235)	× 10 <sup>-7</sup>	× 10 <sup>-7</sup>	× 10 <sup>-7</sup>
11	1	0.1	20	0	45.7	488	+ 5.1	- 17.3	- 12.2
12	1	0.1	20	0.1	45.4	479	+5.1	- 18.9	- 13.8
13	1	0.1	20	0.3	44.9	462	+ 5.3	- 22.4	- 17.1
14	1	0.1	20	1	43.4	418	+ 5.6	- 35.5	- 30.0
15	1	0.1	30	0	52.0	490	+ 3.8	- 22. 2	- 18.4
16	· · 1	0.1	30	0.1	51.7	482	+ 3.8	- 23, 7	-20.0
17	1	0.1	30	0.3	51.2	468	+ 3.9	- 27.1	- 23. 2
18	1	0.1	30	1	49.6	423	+ 4, 2	- 39.6	- 35.5
19	1	0.1	40	0	56.6	481	+ 3, 1	-27.0	-24.0
20	1	0.1	40	0.1	55.5	455	+ 3.2	- 29.8	-26.6
21	1	0.1	40	0.3	55 <b>. 9</b>	462	+ 3.1	- 32.0	-28.8
22	1	0.1	40	1	54.2	422	+ 3.4	- 44. 0	- 40.7
23	1	0.1	50	0	59.3	446	+2.0	- 39.2	- 37.2
24	1	0.1	50	0.1	60.1	465	+1.8	- 38, 3	- 36.5
25	1	0.1	50	0.3	59 <b>.</b> 6	453	+ 1.9	<b>- 41.</b> 5	- 39.7
26	1	0.1	50	1	57.9	387	+2.0	- 53. 5	- 51.5
27	1	0.1	75	0	67.2	438	+1.3	- 48.2	- 46. 9
28	1	0.1	75	0.1	66.9	430	+1.3	- 50.3	- 48.9
29	1	0.1	75	0.3	66.4	421	+1.3	- 52.9	-51.6
30	1	0.1	75	1	64.4	385	+2.1	- 60.1	- 57.9

# TABLE II. SURVEY OF EFFECT OF ADDING SMALL AMOUNTS OF HYDROGEN TO GRAPHITE URANIUM MIXTURES (All compositions contain 10% (by volume) aluminium)

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thermal zone is variable from 0 to 15 cm. The outer reflector consists, for ease of construction, also of water. A natural or depleted uranium buffer or filter separates the fast and thermal zones. The thickness of this filter is 5 cm.





As mentioned earlier, the measurements will have to be performed in several different spectra, thus in more or less diluted compositions of the fast zone. To maintain the fraction of the total reactivity contributed by the fast zone at about the same level (50 to 75%), rather varying dimensions of the fast zone have to be accommodated. To be able to keep the inner radius of the thermal zone at a fixed value when more compact fast zones, which would call for a much smaller radius, are employed, it is suggested that an additional graphite layer of variable thickness should be introduced between the thermal zone and the buffer.

The influence of this additional graphite has been studied in some detail. For a few reference cases the total reactivity is lowered by only 2% when the graphite layer is varied from 0 to 30 cm. This loss in reactivity when compensated by adding fuel to the thermal zone causes a loss in "sensitivity" of 10 to 30%, according to whether a natural or a depleted uranium buffer is being used. For small graphite thicknesses, e. g. 10 cm, even an increase in "sensitivity" is observed. Inspection of the reactivity response versus energy for extreme cases (0 and 25 cm graphite thickness) reveals that over the whole energy range the relative differences are less than 0.75%. This graphite layer of variable thickness thus provides an agreeable freedom in the design.



FIG.9. Perspective view of fast-thermal coupled facility design.



# **BUSTRAAN**

Control plates will be inserted between thermal zone and buffer. Some moderating material (graphite or water) between plates and the thermal fuel zone enhances the control plate values. The actual values are dependent on the reactivity distribution over the thermal zone, buffer and fast zone. Plates covering almost the whole thermal zone may have a total worth of 4 to 6%.



FIG. 11. Vertical cross-section of fast-thermal coupled facility design.

The present design proposal is illustrated in the Figs 9 - 11. This design foresees a central fast zone in a tank of aluminium with an outer diameter of 104 cm and a height of 100 cm, completely separated from the aluminium tank for the thermal zone, thus excluding accidental leakage of water from the thermal tank into the fast tank.

The fast fuel elements will consist of aluminium boxes (1-mm wall thickness), 2 in  $\times$ 2 in in cross-section, in which the platelets of such materials as enriched uranium and graphite are stacked. The platelets will have the same dimensions as used in other facilities, e.g. SNEAK. In the upper and lower side of the elements a 20-cm column of axial blanket material will be located. This will consist of natural or depleted uranium plates possibly alternating with graphite plates. The fuel boxes will rest in a bottom grid. In this gridplate either fuel elements, graphite filler elements or uranium buffer elements may be placed at any location.

The thermal fuel zone consists of 36 thermal fuel elements. These elements, measuring 7.5 cm  $\times$ 15 cm in cross-section, consist of boxes

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in which a variable number of the highly enriched UAl alloy Al-clad plates of 7.5 cm  $\times$ 60 cm can be stacked vertically. The outer section of the thermal tank serves as an outer reflector for the thermal zone. A separation has been made in the bottom part of this outer zone to provide for an air chamber. During the filling of the tank air is trapped in this chamber. The pressure built up can be released by opening magnetic valves. The resulting fast drop of the water level in the thermal zone causes a fast scram in addition to control rod movement.

The existing control rod drive mechanisms of KRITO will be used unaltered. They are located above the system. The oscillator drive system is mounted below the reactor to facilitate the loading of the active samples. In case highly active samples are being oscillated the temperature of the sample will be kept down by a forced circulation of air through the oscillator tube which will remove the decay heat developed by the sample.

A number of gamma-compensated ionization chambers and counters will be located in the outer reflector. From these the neutron flux signals will be derived. For the analysis of the measurements a number of different methods will be used. An experimental auto rod (a small control rod with a servo drive) has been built and is being tested at the Low Flux Reactor at Petten. A direct analysis of the neutron flux signal has already been developed. For reactivity control after loading, extensive use will be made of the pulsed neutron source and reactor noise techniques already developed. The mean neutron life-time will be measured by these. This is a very sensitive safety quantity depending largely on the reactivity contribution of the fast zone. For these purposes a small on-line computer will be installed.

Some data on the three reference compositions in the actual design are given in Tables III and IV.

# 5. PROGRAMME FOR THE MEASUREMENTS AND FOR THE PROCURING OF SAMPLES

After the normal star-up measurements and necessary calibrations, the programme will start with measurements in several spectra on the mock-up fission product material as used in SNEAK. Thereafter measurements in several spectra will be performed on gross fissionproduct samples containing up to 10 to 15 g of fission products of <sup>239</sup>Pu or <sup>235</sup>U, to be produced in the High Flux Reactor at Petten or to be purchased elsewhere. These measurements are intended to produce an over-all figure showing the actual fission product influences compared with the mock-up products. After this a series of more elaborate measurements on chemically separated fission product elements will begin. The amount of sample material needed will vary from a few grams up to 20 g depending on the cross-sections and fission yields of the specific elements or isotopes.

The few fission product elements occurring in nature as a single isotope (as <sup>133</sup>Cs, <sup>139</sup>La, <sup>141</sup>Pr, <sup>103</sup>Rh) will not be difficult to obtain. Other elements, such as Sm, Ru, Nd, Pd, Mo, Eu, Zr, will be separated from actual fission product wastes. A quantity of 500 g of thermal reactor waste has already been ordered for separation purposes.

# BUSTRAAN

		· · · ·	
Composition:	1:30 C	1: 50 C	1:75 C+1 H
Radius fast zone (cm)	30	40	40
Filter natural U (cm)	5	5	5
Thickness graphite (cm)	15	5	5
Thickness thermal zone (cm)	7.4	5	9.7
Extrapolated height (cm)	81.1	81.1	81.1
Mass U-235 (kg)	<u></u>		
Fast zone <sup>a</sup>	120	150	90
Thermal zone	7.5	5	10
Fractional contribution to power			
Fast zone	0.39	0.57	0.28
Filter	0.10	0.11	0.11
Thermal zone	0,51	0.32	0.61
Fractional contribution to reactivity			
Fast zone	0.54	0.75	0.33
Filter	0.11	0.11	0.12
Thermal zone	0.35	0.14	0.55
Mean neutron life-time (µs) <sup>b</sup>	32	12	, 37
Central reactivity value in 10 <sup>-7</sup> per gram fission product			
Absorption	- 16.2	- 23	-23.6
Scattering	+ 2.5	+ 1.2	+ 0.8
Tótal	- 13.7	-21.8	- 22.8
Central reactivity value in 10 <sup>-7</sup> per gram U-235	+28	-	+13

# TABLE III. SOME CALCULATED DATA ON THREE REFERENCE COMPOSITIONS IN THE FAST-THERMAL COUPLED FACILITY

<sup>a</sup> Assuming a saving for an upper and lower blanket of 15 cm each. <sup>b</sup> For pure fast assemblies the life-time would be of the order of  $10^{-6} - 10^{-7}$  s.

	н	С	Al	U <b>-2</b> 35	U-238
1: 30 C	0	6. 38	0.6	0.213	0.0213
1: 50 C	0	6.47	0.6	0.162	0.0162
1: 75 C+1 H	0.087	6.53	0.6	0.087	0.0087

# TABLE IV. ATOMIC DENSITIES (in 10<sup>22</sup> Atoms/cm<sup>3</sup>) OF THE REFERENCE COMPOSITIONS OF TABLE III.

These elements will also be measured in their naturally occurring isotopic composition. Irradiation in the High Flux Reactor of a number of these elements to obtain samples with other isotopic compositions is also being considered. <sup>147</sup>Pm, <sup>135</sup>Cs - <sup>137</sup>Cs and <sup>99</sup>Te will be purchased as sources and purified if necessary. Finally, the purchase or loan of a number of enriched isotopes is being envisaged.

For the chemical operations, a small heavily shielded facility has been designed, in which, by various chemical techniques, for example, ion-exchange methods, the fission products will be separated. In these shielded boxes or in the hot cells of the RCN at the Petten site, the samples will be prepared and mounted for measuring. Sample size and forms will be varied to analyse self-shielding effects.

Besides being used for fission-product cross-section measurements, the system described may be used for measuring cross-sections of other materials or even Doppler coefficients.

According to the present time-schedule we may be able to start the measurements at the end of 1968 or the beginning of 1969. However, negotiations on obtaining and financing the enriched fuel have not yet been concluded. It is estimated that finally up to 200 kg  $^{235}$  U will be needed in this system. In the preliminary periods the amounts needed will be 100 to 150 kg.

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# DISCUSSION

R. VIDAL: The advantage of coupled "thermal-fast" reactors is that the investment in fissile materials can be reduced substantially. Why did you not keep the volume of the fast zone at a minimum? M. BUSTRAAN: From separate calculations for spherical systems it appears that, for small masses, the central and adjoint spectra are far removed from the asymptotic ones. To obtain spectra sufficiently close to the asymptotic ones, one needs, with our isotopic compositions,  $^{235}$ U in amounts of about 100-150 kg for spherical geometries and 180-200 kg for cylindrical geometries.

# ANALYSIS OF INTEGRAL EXPERIMENTS ON THE ZPR VI CRITICAL FACILITY

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# Abstract

ANALYSIS OF INTEGRAL EXPERIMENTS ON THE ZPR VI CRITICAL FACILITY. A new approach is presented to integral experiment evaluation, which correlates nuclear group parameters to the integral data. This correlation is applied to the sodium void experiments made on the ZPR VI critical facility. The basic feature of the method consists in the use of generalized perturbation techniques and in the minimization of nuclear parameter corrections.

# 1. INTRODUCTION

At the present time, much effort is being given in fast reactor physics to the correlation methods between measured and calculated integral quantities, considered as necessary complements to basic data evaluation. The main reason for this interest may be summarized as follows:

- A higher degree of accuracy than that existing at present in basic nuclear data seems to become increasingly difficult to achieve, time consuming and expensive, although there is a need for more accurate theoretical predictions.
- (2) What reactor designers are most concerned with is not the basic data in themselves, but the integral behaviour of the reactor or, quantitatively, reaction rates, neutron lifetimes, reactivity worths, etc.
- (3) A large amount of integral information is becoming increasingly available from fast critical facilities and/or new fast prototypes.
- (4) Larger and faster computing facilities are being installed.

Naturally these reasons call for a compromise of efforts between basic and integral data evaluation work. There is no doubt that each of these activities will eventually mutually benefit.

The scope of the present paper is to give a general description of the philosophy of the correlation method adopted, and to show some results relevant to the ZPR VI critical experiments.

# 2. GENERAL PHILOSOPHY OF THE METHOD

The basic philosophy of the method is simple and may be found in Ref.[1]. It will be described here very briefly together with some recent developments. Let us consider a number I of integral experiments of the most general kind. They may belong to one or more reactor systems.

and may represent in general reaction rate ratios, reactivity worths, reactivity ratios, prompt neutron lifetimes, and source worth ratios.

We shall first reduce the integral quantities according to the simplified calculational models assumed for each experiment, in order to account for heterogeneity, shape, self-shielding and other effects. We shall then associate with all these experiments an I-component vector  $(\vec{m} + \vec{m})$  where  $\vec{m}$  represents the experimental reduced integral data and  $\vec{m}$ ' represents the inaccuracies connected with them in relation to both the statistics of the experiments and the data reduction procedure from microscopic to group cross-sections. Let us now assume by best value criteria (the basis is a fiduciary approach) J-multigroup nuclear constants. The multigroup approximation is consistent with the fact that it is widely used for fast reactor calculations, and therefore it represents the form by which the cross-sections are required. We further assume that the iterative method used by Abagyan et al. [2] is valid. With this assumption the J-group constants will be normalized to infinite dilution. The selfshielding factors will not be affected by the present method, at least at the stage it is intended at present. The J-multigroup constants will represent in general group averages defined in accordance with the theoretical approximation used. This is described in detail in Ref. [2]. These constants will represent generally scattering, absorption, fission cross-sections, number of secondaries per fission, and fission spectra. They will be represented by a J-component vector  $\vec{d} + \vec{d'}$  where  $\vec{d}$  represents the infinite dilution group constants and  $\vec{d'}$  the inaccuracies connected with them. The same arguments used for  $\vec{m}$ ' are applicable here. It is possible, in particular, that the group reduction of basic data introduces some degree of uncertainty which may be accounted for by properly adjusting  $\vec{d'}$ .

If we now calculate the I-integral quantities corresponding to the measured ones, by making use of the J-group constants  $\vec{d}$  and adopting given theoretical models we shall generally observe discrepancies  $\Delta \vec{m}$  between the measured and calculated values. Let us suppose then that we are able to express  $\Delta \vec{m}$  in terms of cross-section adjustments  $\Delta \vec{d}$  in a simple linear form of the type

$$\Delta \vec{m} + \epsilon \vec{m'} = L \Delta \vec{d} \tag{1}$$

where  $\epsilon$  is an (I×I) diagonal matrix in which the elements  $\epsilon_i$  are undetermined parameters with limits  $|\epsilon_i| \leq 1$  and where L is an (I×J) matrix in which the elements  $a_{ij}$  represent calculable sensitivity coefficients. This is allowed for if use is made of generalized perturbation techniques shown in Refs [3-5]. At this point a consistency approach should be applied in correcting the group constants by amounts  $\Delta \vec{d}$  within their uncertainty limits  $\vec{d}'$  so that agreement is obtained between theoretical and experimental values within their uncertainty limits  $\vec{m}'$ . If the problem has solutions, since we assume that in general I < J, there is an infinite number of constant sets consistent with the experimental values. To arrive at a unique set, a least norm condition is applied to corrections  $\Delta \vec{d}$ . More precisely the following condition must be satisfied:

$$F = \Delta \vec{d} A^{-1} \Delta \vec{d} = minimum$$

(-)

where  $A^{-1}$  is the following diagonal matrix:

$$A^{-1} = \operatorname{diag}\left(\frac{1}{\alpha_1^2}, \frac{1}{\alpha_2^2}, \dots, \frac{1}{\alpha_1^2}\right)$$

where  $\alpha_j$  is a parameter usually assumed proportional to the absolute error d'<sub>j</sub>. This assumption stems from the principle of greater correction for those constants which look affected by higher inaccuracies. It may appear that some corrections  $\Delta d_j$  result from the bounds represented by their uncertainty limits. In such case, the pertinent upper or lower bounds should be assumed as the proper correction and there should be a new iteration with d'<sub>j</sub> (and therefore  $\alpha_j$ ) close to zero. This process may be iteratively applied until, it is hoped, convergence to consistency is reached. If this is the case, we may say that the method adopted, although not strictly based on physical grounds, is, however, operating within the uncertainty area of what can be assumed to be the present knowledge of <u>both</u> the differential and integral parameters and gives "effective" group constants consistent with it.

It may happen, however, that convergence is not obtainable either because the errors assumed for the group constants and/or integral experiments are unreal and/or because the calculational models and approximations are inappropriate. At this point assessment in terms of general good sense is essential, as some sort of judgement on basic and integral data and/or theoretical assumptions will be implied. Some kind of sensitivity test might be used to help this situation, by slightly changing the uncertainty limits  $\vec{m}'$  of the experimental integral data, in order to sort out those data which appear most critical for the convergence to consistency and therefore are worth looking at first.

Mention should be made of the fact that the method outlined here may be used also for checking pieces of information assumed to be reliable with others assumed uncertain. For example, one might want first to force consistency between given group constants and a given set of reliable experimental integral data, and then to check a new set of similar experimental values with the corrected cross-sections. This also represents a possible way of sorting-out those pieces of information which might cause trouble.

A final comment is required in relation to possible intercorrelations among cross-sections. In principle, system (1) can accept different linear equations. Therefore, the system of equations representing these intercorrelations given with obvious notation by the expression

$$\vec{q} + \epsilon_1 \vec{q'} = L_1 \Delta \vec{d}$$

may be directly incorporated by it. If it happens, which is unlikely, that I > J, a fit procedure may be applied, so that

$$\mathbf{F}_1 = (\Delta \vec{m} - L \Delta \vec{d}) \mathbf{B}^{-1} (\Delta \vec{m} - L \Delta \vec{d}) = \text{minimum}$$

where  $B^{-1}$  is the following diagonal matrix:

$$B^{-1} = diag\left[\frac{1}{(m'_1)^2}, \frac{1}{(m'_2)^2}, \dots, \frac{1}{(m'_1)^2}\right]$$

	Na				235 <sub>U</sub>				238 <sub>U</sub>		F	e	с
Group	♂el	$\sigma_{in}$	σ <sub>c</sub>	ν	σf	a <sub>in</sub>	σ <sub>c</sub>	ν	σ <sub>f</sub>	Jin	<sup>o</sup> el	o <sub>in</sub>	a <sub>el</sub>
1	1,62	0,65	0.0286	3,08	1.234	1,909	0.0086	3.14	0.629	2.521	2,385	1,352	1,297
2	1,95	0,65	0.04	2.79	1.25	1.91	0.02	2.87	0.58	2,60	2,37	1.13	2.00
3	2.42	0.58	0.06	2.63	1.28	1.76	0.06	2.67	0,49	2.25	2.40	0.90	1.80
4	3,32	0.48	0.12	2,52	1.25	1.38	0,13	2,58	0.02	2.15	2,53	0.37	2,55
5	4.37	0.13	0,17	2.46	1,23	1.20	0.13	0.0	0.0	1.65	3,78	0.01	3,10
6	4.00	0.0	0.25	2.47	1.41	1.00	0,15			1,05	2,99	0.0	4.0
7	3.80		0.40	2.45	1.70	0.60	0.22			0,55	3,69		4,3
8	5.30		0.60	2.44	2.10	0.18	0.35			0.19	5.29		4.5
9	4.30	÷	1.00	2.43	2.65	0.06	0.46			0.0	14.5		4.6
10	5.00		1,50	2.42	3.40	0.0	0.60				3.99		4.7
11	8.00		2.10	2.42	4.40		0.78				8.40		4.7
12	100.0		2.75	2,42	5.40		1.20				5,89		4.7
13	6.2		3.80	2,42	7.30		2,10				7.39		4.7
14	3,3		6.30	2.42	11.0		3,60				10.0		4.7
15	3.1		9.50	2,42	16.0		4,50				11.0		4.7
16	3.1		15,11	2.42	24.31		17.64				11.4		4.7

# TABLE I. 16-GROUP CROSS-SECTIONS FROM THE SOVIET LIBRARY (barns)

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The J-conditions  $\delta F_1/\delta d_j$  give the J-equations from which the corrected set may be deduced.

# 3. CALCULATIONAL PROCEDURE

To solve system (1) the CALI code has been used [6]. The sensitivity coefficients  $\alpha_{ij}$  of matrix L are now calculated separately making use of two separate codes, the CIAP-2D and the GLOBPERT-2D. The CIAP-2D [7] calculates in two dimensions and multigroup diffusion theory the global importance and real flux functions used in the generalized perturbation techniques. In input, it requires on tape real and adjoint flux maps from a bi-dimensional diffusion code [8]. The GLOBPERT-2D code [9] uses the fluxes of the CIAP-2D code, and calculates the coefficients  $\alpha_{ij}$  using procedures analogous to those adopted in conventional perturbation techniques.

To reduce the experimental data to the homogeneous calculational model adopted, the heterogeneity effects have been evaluated by using a first collision probability method [10]. These effects, however, have been shown to be negligible in the cases considered. As the best set of basic values, the library of Abagyan et al. [2] has been adopted with proper consideration given to the self-shielding factors [11]. To reduce the calculation time, a reduction of the energy groups from 26 to 16 has been made. This has been obtained by collapsing in one group the first two and the last ten energy groups of the Soviet library. With the same aim to save computing time, a pre-selection of nuclear parameters significantly relevant to the integral experiments considered has been made. This pre-selection has been based on a previous sensitivity analysis. The nuclear parameters which have resulted are:  $\nu$ ,  $\sigma_f$ ,  $\sigma_c$ ,  $\sigma_{in}$  of <sup>238</sup>U,  $\sigma_{el}$ ,  $\sigma_{in}$  of Fe and  $\sigma_{el}$  of C. Their initial values are shown in Table I.

# 4. APPLICATION OF THE METHOD TO ZPR VI INTEGRAL EXPERIMENTS

As a starting point in using the correlation method outlined above, two sodium removal experiments of Assembly 2 of ZPR VI [12] have been chosen, considering the general relevance of this kind of experiment on the sodium void effect. The material compositions and dimensions of this assembly are shown in Table II. In Table III the regions where the sodium has been voided in integral experiments under study in the present analysis are indicated together with the experimental and calculated values. As they are related to the inner and outer core regions, these experiments should give evidence of the spectral effects and the leakage effects, and therefore should be most significant. As may be seen, there is an excellent agreement, of  $\sim 1\%$ , for the central removal experiment and a good agreement, of  $\sim 10\%$ , for the eccentric removal experiment. If we consider also the experimental errors assumed, there is evidence of the rather good reliability of the Soviet cross-section library adopted, and of the iteration method implied in it for evaluating the self-shielding factors. The use of the CALI code has given, as expected, small variations, of the order of a few percent, in the scattering TABLE II. DIMENSIONS (cm) OF ASSEMBLY 2 OF ZPR VI AND COMPOSITIONS (nuclei/cm<sup>3</sup> × 10<sup>-24</sup>)

	R	H <sup>a</sup>	235 <sub>U</sub>	238U	С	Na	SS
Core	46,22	45,72	0.002285	0,009853	0.0129	0.00791	0.01475
Blanket	76.72	76.22	0.000081	0,03993	0.0	0.0	0.00965 <sup>b</sup> 0.006003

<sup>a</sup> From midplane.
 <sup>b</sup> The two values refer to the inner and outer part of the blanket.

#### Na VOIDED REGIONS: EXPERIMENTAL AND CALCULATED VALUES (in $\Delta k/k \times 10^{-5}$ ) TABLE III.

	R <sub>in</sub> (cm)	R <sub>out</sub> (cm)	H <sub>in</sub> (cm)	H <sub>out</sub> (cm)	Reduced exp. value	Calculated value
Integral experiment B	0,0	13.592	0.0	15.24	- 2.62±0.13	- 2.60
Integral experiment I, III	24, 468	35,344	35.56	45.72	-137.4 ±7.8	-120.0

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	N	a .		235	U			238U			Fe		. c
Group	<sup>o</sup> el	ơin	σ <sub>c</sub>	ν	σ <sub>f</sub>	o <sub>in</sub>	σ <sub>c</sub>	ν	af	ơ <sub>in</sub>	<sup>a</sup> el	o <sub>in</sub>	<sup>o</sup> el
1	0,198	0.177	0.0	-0.004	-0.006	0.056	0.002	-0,010	-0.001	0.322	0.030	0.019	0.053
2	0.459	0.152	0.001	-0,002	-0.004	0.078	0.006	-0.003	-0.005	0.466	0.081	0.075	0.144
3	1,137	0,500	0.001	-0.003	-0.006	0,085	0.023	-0,004	-0.006	0.544	0.223	0,362	0.337
4	2,196	0.415	0.001	-0.004	-0.008	-0,045	0,023	-0.001	-0.001	-0,009	0,317	0.285	0.344
5	4,432	-0.008	0.004	-0.010	-0.018	-0,136	0.014	0.0	0.0	-0.082	0.399	0,551	-0.273
6	2.081	0.0	0.012	-0.004	-0.007	-0.011	0.032			-0.024	0.196	0.0	-0,034
7	1.078		0.014	0.001	0.002	-0.003	0.033			0,002	0,102		0,063
8	1.007		0.004	0.006	0.012	-0.001	0.011			-0.006	0.046		-0.021
9	0,550		-0.019	0,012	0.023	0.0	-0.038			0.0	0.041		0.025
10	0.414		-0.035	0.013	0.024	0.0	-0,056				0,023		0.066
11	0.267		-0.035	0.009	0.018		-0.048				0.049		0.066
12	1,112		-0,003	0.0	0.001		-0.004				-0.004		0.031
13	0.115		-0.055	0.011	0.022		-0.056				0, 021		0.035
14	0.020		-0.038	0.006	0.012		-0.029				0.013		0.024
15	0.001		-0.019	0.002	0.004		-0.015				0.006		0.013
16	-0.002		-0.018	0.001	0.002		-0.012		·		0.0		-0.001

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# TABLE V. EXPERIMENTAL AND CALCULATED VALUES (in $\Delta k/k \times 10^{-5}$ ) WITH CORRECTED CROSS-SECTIONS

	R <sub>in</sub> (cm)	R <sub>out</sub> (cm)	H <sub>in</sub> (cm)	H <sub>out</sub> (cm)	Reduced exp. value	Calculated value
Integra1 experiment C	0.0	19,030	0.0	20.32	- 92.8±4.6	- 95.6
Integral experiment I, I	24.468	35.344	0.0	10.16	-166.1±8.3	-168,5
Integral experiment I, II	24.468	35.344	17.78	27.94	-153.6±7.7	-150.7
Integral experiment J, III	. 35,344	46.220	35.56	45.72	-136.3±6.8	-132.9

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cross-sections of sodium for which the eccentric measurement appears by far the most sensible. All these corrections, as assumed, lie well within the cross-section uncertainties, and are shown in Table IV.

To check the validity of the "effective" cross-sections obtained, they have been used to calculate the other sodium void experiments of Assembly 2. The agreement has proved excellent, as shown in Table V.

The next step in this work will be the analysis of the zoned sodium void experiments of Assembly 4Z of the ZPR VI critical facility. They will first be interpreted in terms of corrected cross-sections (and realistic geometries) and, if there is no agreement, they will be assimilated by the correlation procedure adopted.

# ACKNOWLEDGEMENT

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# DISCUSSION

J.Y. BARRE: I do not feel that one is justified in correcting the cross-sections for five elements and 16 energy groups (Table IV) on the basis of two series of measurements (Table III).

G. CECCHINI: That may be so. However the results obtained with the "corrected" cross-sections are very good.

J.Y. BARRE: The small number of measured parameters you use imposes an additional constraint on the cross-sections, so that you have an over-determined system. Like Mr. Pendlebury, with the PENICUIK code, and Mr. Yeivin, with the method he presented in paper SM-101/67, your method requires that the sum of the squares of the cross-section variations be minimized. This is equivalent to assuming that your initial cross-sections are exact or that there are no longer any systematic errors in the cross-sections.

G. CECCHINI: Our method requires that the functional F be minimized, not the sum of the squares of the cross-section variations. Suitable values of  $\alpha_j$  may be selected when considering the systematic errors.

Y. YEIVIN: With regard to Mr. Barré's remark, I should like to point out that there is no simple direct relationship between the amount of integral data and the extent to which cross-sections (or group constants) may be modified. The results I presented were obtained by employing critical mass data for 18 systems; they could also have been obtained, however, by using as few as three systems. Adding more systems of the same kind does not yield new cross-section information; only the addition of different systems (different from, say, the spectrum point of view) will contribute significantly to the cross-section modifying procedure.

J.L. ROWLANDS: It seems to me that the accuracy of the predictions made using adjusted cross-sections is not so apparent as when a simple adjustment factor (to be applied to the calculated parameter) is derived from the experiments. Have you considered assessing the accuracy of your adjusted cross-sections or of the predictions made with them?

G. CECCHINI: Yes, and, as I said, we found that the values calculated using the corrected cross-sections are in very good agreement with the experimental data.

K.H.JOEST: If I have understood your paper correctly, your correction of group constants consists in a modification of infinitely dilute values. However, the self-shielding factors may also be incorrect, and this may be of importance, for instance, in fitting measured Doppler coefficients. Can you also handle such problems with your method?

G. CECCHINI: Yes, we can.

# THEORETICAL AND EXPERIMENTAL ANALYSIS OF FAST NEUTRON SPECTRA

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#### Abstract

THEORETICAL AND EXPERIMENTAL ANALYSIS OF FAST NEUTRON SPECTRA. The reactor physics division of the Inter-Academic Reactor Institute at Delft is concentrating its efforts in the field of fast reactor physics on problems of a more fundamental nature. The object of the programme is to determine experimentally a number of microscopic reactor physics parameters such as conversion potentials, fission ratios and Doppler coefficients for simple geometries and material compositions. Because of the extreme importance of knowledge of the neutron spectrum for the interpretation of the results, attention has initially been concentrated on both the measurement and the calculation of fast neutron spectra.

The transport of neutrons in absorbing and non-absorbing heavy atom materials is studied by solving the Boltzmann equation. Both isotropic and anisotropic scattering are considered. Anisotropic scattering is treated by the  $P_n$ -approximation, while flux-anisotropy is handled with the  $S_N$ -method. In the code FAST-DELFT, scattering is treated up to the  $P_4$  component, a further extension of which is useless because of the lack of available cross-section data. By using this method, the effect of scattering anisotropy on the spectrum formation has been studied. In addition the influence of group cross-section inaccuracies was determined. The experimental work has been concentrated on methods to determine in-core spectra. Using home-made proportional counters with gamma-ray discrimination provisions fast neutron spectra have been measured in simple geometries. These experiments were complemented by foil measurements in the lower energy region. The results of this work are presented in this paper.

# 1. INTRODUCTION

The reactor physics division of the Reactor Instituut at Delft is concentrating on some fundamental problems in fast reactor physics.

The object of the programme is to determine experimentally several microscopic reactor parameters such as conversion potentials, fission ratios and Doppler coefficients for simple geometries and material compositions in order to test the validity of calculation methods and possibly improve them.

We feel that a university laboratory may thus, to a certain extent, contribute to a better understanding of the neutron behaviour in fast reactors, at the same time confronting students, majoring in reactor physics, with current developments.

Because knowledge of the neutron spectrum is essential for interpreting the experimental results, attention has been first focused on both the measurement and the calculation of fast neutron spectra.

Theoretical work involves the analysis of neutron transport in various materials by solving the Boltzmann equation using the diffusion approximation [1] and the more rigorous  $S_N$  method [2,3]. In particular the influence of anisotropic scattering on the space and energy distribution of neutrons is being investigated.

Two different transport approximations to correct for anisotropic scattering are compared (diagonal transport approximation and a corrected diagonal transport approximation), which can be used in codes dealing with only isotropic scattering. The adequacy of these approximations will be investigated by comparing the results with those of an anisotropic version of the  $S_{\rm N}$  code, based on a  $P_{\rm n}$  treatment of anisotropic scattering.

The experimental work has concentrated on methods to determine incore spectra. Using foil activation methods and home-made proportional counters, fast neutron spectra are being measured in simple geometries with relatively small volumes. In addition to rise time discrimination [4], a well-known method for eliminating the counter gamma-response, a technique is being developed for drastically reducing the gamma sensitivity by applying, inside the counter tube, an anti-coincidence shield against conversion electrons originating from the wall.

In section 2 of this paper an outline is given of the transport approximations while a more detailed presentation of the experimental work appears in section 3. The calculation results are presented in section 4.

# 2. THEORETICAL ANALYSIS

# 2.1. Transport approximations for anisotropic scattering

A transport approximation, additional to the commonly used diagonal transport approximation (DTA) as described by Pendlebury and Underhill [5], was introduced for use in an isotropic scattering version of the transport equation to replace a more rigorous calculation model that includes anisotropic scattering.

For demonstration purposes a non-multiplying medium with a slab geometry is considered. The one-dimensional transport equation for this case reads:

$$\mu \frac{\mathrm{d}}{\mathrm{d}x} \mathrm{N}(\mathrm{x}, \mathrm{E}, \mu) + \Sigma_{\mathrm{t}}(\mathrm{x}, \mathrm{E}) \mathrm{N}(\mathrm{x}, \mathrm{E}, \mu) = \mathrm{S}(\mathrm{x}, \mathrm{E}, \mu) \tag{1}$$

where: x

μ

= space co-ordinate

= cosine of the angle between the abscissa and the flight direction of the neutron

 $N(x, E, \mu)$  = flux of neutrons with energy E per unit  $\mu$ -interval about the direction  $\mu$  at x

 $S(x, \dot{E}, \mu)$  = source of neutrons with energy E per unit  $\mu$ -interval about the direction  $\mu$  at x, containing scattering sources and external, i.e. flux-independent sources.

Since external source contributions are not essential for the present discussion, further reference to them is omitted.

Expanding fluxes and scattering cross-sections in Legendre-series, retaining terms up to the  $P_1$  component and discretizing the energy variable, yields multigroup  $P_1$  equations for neutron transport.

$$\frac{\mathrm{d}}{\mathrm{d}\mathbf{x}} \phi_1^{\mathrm{g}} + \Sigma_t^{\mathrm{g}} \phi_0^{\mathrm{g}} = \sum_{\mathrm{g'}} \Sigma_0^{\mathrm{g'g}} \phi_0^{\mathrm{g'}}$$

$$\frac{1}{3} \frac{\mathrm{d}}{\mathrm{dx}} \phi_0^{\mathrm{g}} + \Sigma_t^{\mathrm{g}} \phi_1^{\mathrm{g}} = \sum_{\mathbf{g}'} \Sigma_1^{\mathrm{g}'\mathrm{g}} \phi_1^{\mathrm{g}'}$$

where:

 $\Sigma_{t}^{g}$ ,  $\Sigma_{1}^{g'g}$ 

 $\phi_{\alpha}^{g}, \phi_{1}^{g}$ 

= zero and first-order, respectively, Legendre component of the flux in energy group g = total cross-section for neutrons in energy group g = zero and first-order, respectively, Legendre component of the cross-section for scattering of neutrons from group g' to group g.

As shown by Pendlebury and Underhill [5], a transport approximation is obtained by introducing the following substitutions in Eqs (2a) and (2b):

$$\Sigma_{tr}^{g'g} = \Sigma_{t}^{g'g} \delta^{g'g} - \Sigma_{1}^{g'g}$$
(3a)  
$$\Sigma_{str}^{g'g} = \Sigma_{0}^{g'g} - \Sigma_{1}^{g'g}$$
(3b)

$$\delta^{g'g} = 0$$
 if  $g' \neq g$ , or  $\delta^{g'g} = 1$  if  $g' = g$ 

As a result the terms with  $\Sigma_t^g \phi^g$  in Eqs (2a) and (2b) must be replaced by summations of g'-dependent terms. These modifications, known as the full transport approximation, require the calculation of the firstorder Legendre components of the group fluxes, and therefore cannot be treated with codes based on isotropic scattering sources.

When the transport cross-sections, as given in Eqs (3a) and (3b), are used in a code that handles flux anisotropy more rigorously than that up to the  $P_1$  component (e.g.  $S_4$  or  $S_8$  approximations) it can be shown that the transport approximation becomes exact in two situations: (a) For the trivial case where the  $\ell$ -th order scattering cross-section Legendre component  $\Sigma_{\ell}^{g\,g} = 0$  for  $\ell > 0$ , i.e. isotropic scattering. (b) When  $\Sigma_{\ell}^{g\,g} = \overline{\mu}^{g\,g} \Sigma_{0}^{g\,g}$  for  $\ell > 0$ , where  $\overline{\mu}^{g\,g}$  is the mean cosine of the scattering angle in the laboratory co-ordinate system for scattering from energy group g' to g.

In the latter case, where all higher-order Legendre components of the scattering cross-section are equal, it can easily be shown that the angle-dependent scattering cross-section can be written as:

$$\Sigma_{s}(\mu) = \frac{\Sigma_{s}}{2} \left\{ 1 - \overline{\mu} + 2\overline{\mu} \cdot \delta(1 - \mu) \right\}$$
(4)

where  $\delta$  is the Dirac delta function.

Physically this means that scattering consists of an isotropic component plus a purely forwardly directed component.

However, the real advantage, a decrease in machine time, will be obtained when transport approximations can be incorporated in codes dealing with isotropic scattering sources.

(2b)

(3b)

A well-known method is to ignore scattering anisotropy in the slowingdown cross-sections. This method is referred to as the diagonal transport approximation (DTA) [5]. In this case the cross-sections are given by:

$$\Sigma_{\rm tr}^{\rm g} = \Sigma_{\rm t}^{\rm g} - \Sigma_{\rm l}^{\rm gg} \tag{5a}$$

$$\sum_{str}^{g'g} = \sum_{0}^{g'g} - \sum_{1}^{g'g} \cdot \delta^{g'g}$$
(5b)

When an appreciable part of the slowing down in high-energy regions is due to elastic scattering – as is the case for systems containing a certain amount of light- to medium-weight nuclides – the DTA is expected to give less accurate results. For these conditions we use a corrected diagonal transport approximation (CDTA). This correction is obtained by substituting cross-sections in such a way that the distribution of scattering angles for those neutrons that remain in the energy group considered, is taken into account so that the mean cosine of the total number of scattering events equals the actual mean cosine of the angular distribution, i.e.:

$$\Sigma_{tr}^{g} = \Sigma_{t}^{g} - \sum_{g'} \Sigma_{1}^{gg'}$$

$$\Sigma_{str}^{g'g} = \Sigma_{0}^{g'g} - \delta_{0}^{g'g} \sum_{g''} \Sigma_{1}^{gg''}$$
(6a)
(6b)

The DTA and CDTA, as given by Eqs (5) and (6), are in fact special cases of the full transport approximation as can be illustrated by considering the event where the group width is such that elastic scattering of neutrons can only extend to the nearest lower energy group. The equations describing this condition are:

$$\frac{d\phi_1^8}{dx} + \Sigma_t^g \phi_0^g = \Sigma_0^{gg} \phi_0^g + \Sigma_{er,0}^{g-1} \phi_0^{g-1}$$
(7a)

$$\frac{1}{3} \frac{\mathrm{d}\phi_0^g}{\mathrm{d}x} + \Sigma_t^g \phi_1^g = \Sigma_1^{gg} \phi_1^g + \Sigma_{\mathrm{er},1}^{g^{-1}} \phi_1^{g^{-1}}$$
(7b)

where:  $\Sigma_{er,0}^{g-1}$ ,  $\Sigma_{er,1}^{g-1}$  = zero and first-order, respectively, Legendre component of the cross-section for elastic transfer of neutrons from group g-1 to group g. This crosssection is frequently referred to as the elastic reduction cross-section.

Eq.(7b) can be modified to:

$$\phi_1^{g} = -\frac{1}{3\left(\Sigma_t^{g} - \Sigma_1^{gg} - \Sigma_{er,1}^{g-1} \frac{\phi_1^{g-1}}{\phi_1^{g}}\right)}\frac{d\phi_0^{g}}{dx}$$

(7c)

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To eliminate the  $P_1$  component in the source term of the transport equation, either of the following two approximations can be introduced in Eq.(7c).

(i)  $\Sigma_{er,1}^{g-1} \phi_1^{g-1} = 0$ , i.e. anisotropic slowing down is neglected (DTA)

or

(ii)

 $\Sigma_{er,1}^{g^{-1}} \phi_1^{g^{-1}} = \Sigma_{er,1}^g \phi_1^g$ , i.e. anisotropic scattering into the group considered is equal to anisotropic scattering from the group (CDTA).

Physically this involves the assumption that, should the second approximation (CDTA) be used, all anisotropic scattering is to be intragroup scattering, whereas in the DTA case only the intragroup contribution to anisotropic scattering is accounted for in the transport equation.

The net effect of these approaches is that scattering is less anisotropic for the CDTA case than for the DTA case, because the forwardly directed anisotropy of slowing-down collisions is less than that for the intragroup collisions, owing to larger energy losses associated with high angle collisions. Since in both approximations slowingdown scattering is treated as being isotropic, the total scattering angular distribution (isotropic+delta forward) is more accurately accounted for in the CDTA case.

At present the results of calculations based on the CDTA case are being compared with those of  $\rm S_N$  calculations, including anisotropic scattering, to determine the limits of applicability of these approximations.

# 2.2. Group cross-sections

The ABN-set [6] has been used for calculations analysing the experimental arrangement as described in section 3. In this set a standard spectrum was used for averaging cross-sections over energy-groups consisting of the fission spectrum for three groups above 2.5 MeV and a Fermi spectrum (1/E) for all lower energy-groups. For the systems being studied discrepancies are encountered owing to differences between the standard spectrum and the actual spectrum: (i) In the case of graphite, discrepancies are expected near the fission source where the spectrum deviates strongly from the Fermi spectrum. However, at some distance the group cross-sections will be correct. (ii) In the case of aluminium it is necessary to develop a new cross-section set because of the great discrepancy between the ABN-spectrum and the actual spectrum in all positions of the lattice.

A computer code for the calculation of cross-sections for groups with resonances is being developed. It is based on the same principles as ELMOE [7]. An additional problem to be solved is that no asymptotic spectrum exists in the system so that group cross-sections will be space-dependent. Until now the self-shielding factors as listed in the ABN-set were used.

A preliminary study has been made of the deviations from ABN cross-sections caused by the discrepancy between the actual spectrum and the Fermi spectrum. Elastic transfer cross-sections are most sensitive to these deviations. Since the actual spectrum is harder than the Fermi spectrum, elastic transfer cross-sections will be smaller than those given by the ABN, resulting in harder spectra than obtained with ABN cross-sections. Writing the intragroup flux of lethargy u as  $\phi(u) = e^{-Bu}$ , where B = 0 represents a Fermi spectrum, the elastic reduction cross-section  $\Sigma_{er}$  for the group from lethargy u<sub>1</sub> to lethargy u<sub>2</sub> is given by:

$$\Sigma_{er} \equiv \int_{u_1}^{u_2} e^{-Bu} du = \int_{u_1}^{u_2} \Sigma_{b} (u \rightarrow u' > u_2) e^{-Bu} du$$
(8)

By further assuming the total scattering cross-section to be constant within the group one finds:

$$\Sigma_{\rm er} = \frac{\Sigma_{\rm s}}{1-\alpha} \quad \frac{B}{e^{\rm B}\Delta u_{-1}} \left\{ \frac{1-e^{(1-B)\ln\alpha}}{1-B} + \frac{\alpha}{B} \left(1-e^{-B\ln\alpha}\right) \right\}$$
(9)

where:  $1-\alpha$  = maximum fractional energy decrease per elastic collision,  $\Delta u$  = group width.

For B = 0 Eq. (9) gives the expression used for the ABN-set:

$$\Sigma_{\rm er} = \frac{\xi}{\Delta u} \Sigma_{\rm s} \tag{10}$$

where  $\xi$  = mean logarithmic energy decrement per elastic collision.

Table I shows the decrease of the elastic reduction cross-section as a function of B for the values  $\Delta u$  in the ABN-set.

TABLE I. DECREASE OF ELASTIC REDUCTION CROSS-SECTION (%) AS A FUNCTION OF B AND  $\Delta u$  RELATIVE TO THE VALUE FOR B = 0

В	Lethargy interval							
	$\Delta u = 0.57$	∆u = 0.69	$\Delta u = 0.77$					
0.1	1.1	1.7	2.1					
0.2	2.9	4.1	4.9					
0.3	4.6	6.4	7.5					
0.4	6.4	8.7	10,2					
0.5	8.5	10.9	12.8					

For graphite the results of the calculations show that near the fission source B varies between 0.5 and 0.1 for the groups 5 to 14 (1.4 MeV to 1 keV). This means that the elastic reduction cross-section may deviate up to 11% from the values given by ABN. The result of a re-evaluation of cross-sections in the region near the source has been investigated (see section 4).

# 3. EXPERIMENTAL WORK

The experimental programme requires a facility in which various neutron spectra can be created. To compare experimental results with those of calculations this facility should comprise simple material compositions and geometries.



FIG.1. Vertical cross-section of the HOR-university pool reactor thermal column.

In the centre of a graphite block  $(110 \times 110 \times 110 \text{ cm}^3)$  on top of the thermal column of the 2-MW HOR-university pool reactor (see Fig. 1) different materials of interest can be stacked together with fission plates to create neutron energy distributions that are strongly affected by anisotropic scattering.

Provisions have been made for the insertion of counter tubes and foils for spectrum measurements at different positions in the lattice. The geometrical arrangement of the materials has been selected so that onedimensional calculations for a spherical geometry seem to be an acceptable approximation for the system proper.

To calibrate the equipment and to test calculation methods the system was first uniformly filled with graphite. Thus the spectrum was properly defined and no difficulties arose in defining the group constants.

As a material of primary interest aluminium will be used, which exhibits many strong scattering resonances in the 10 keV-1000 keV energy range. In particular, attention will be paid to the spectral behaviour near the aluminium-graphite interface. To introduce changes in the spectrum gradient various concentrations of light nuclei will be mixed with the aluminium. Thereafter these materials will be partly replaced with fissionable materials.



FIG.2. Radial cross-section of the anti-coincidence counter tube.

The neutron spectra are being measured by both activation methods and proton recoil counters. As a possible means of reducing the gamma response of a proton recoil tube a special technique was developed for discrimination between conversion electrons from the wall and those generated in the gas filling. Figure 2 gives a radial cross-section of this counter. It consists of a centrally located main anode surrounded by 16 cathode wires dividing the volume into an internal and an external zone. Ionizations created in the external volume are detected by eight guard anodes. By means of anti-coincidence operation the contribution of wall conversion electrons is eliminated. Because the main component of the gamma response is caused by this effect a considerable decrease of gamma sensitivity is expected to be obtained. A further advantage of this design is that only those protons are detected that transfer their total energy in the central region of the tube. This simplifies correction procedures. A minor disadvantage is a decrease in yield. End-effects still exist but are of minor importance because the two axial surfaces make up only about 14% of the total surface for a sensitive volume 2-in. long and  $\frac{2}{3}$  in. in diam., as is used in our case.

# 4. CALCULATIONAL RESULTS

Spectrum calculations were performed for a spherical geometry of the following dimensions:

r = 0 - 6 cm (central hole (vacuum))
r = 6 - 7 cm (shell shaped fission source)
r = 6 - 30 cm (first zone (graphite or aluminium))
r = 30 - 55 cm (second zone (graphite)).
Figure 3 shows spectra in a graphite system, calculated with diffusion and  $S_4$  approximations. In the central hole a relatively hard spectrum exists whereas, at a distance of 26 cm from the centre, the spectrum is almost 1/E below 400 keV. A comparison between diffusion and  $S_4$ approximations shows that the flux level calculated with diffusion theory is too low, especially near the source, because first-flight neutrons are not accounted for. The influence of this effect is, of course, stronger with increasing removal to the intragroup scattering ratio. For the lower energy groups the discrepancy is smaller because, for these groups, the scattering sources are more spatially distributed.



FIG.3. Spectra in a pure graphite system.

Results of a comparison between the two transport approximations developed in section 2 are shown in Fig.4. The CDTA gives a softer spectrum than DTA owing to the stronger anisotropy that is assumed in the DTA, as was indicated earlier. The calculations show that group fluxes differ up to 10% in the lower energy range whereas, in the case of DTA, the leakage from the system is approximately 16% higher. As mentioned in the introduction, more specific conclusions about the validity of these transport approximations can be given by comparison with more rigorous calculations.

The influence of a modification of elastic transfer cross-sections, according to section 2, was studied by using modified cross-sections for the region between r = 6 and r = 26 cm and the original ABN-set for the remaining part of the system. The results of these calculations, indicating a hardening of the spectrum, are presented in Fig.5.



FIG.4. Relative differences between group fluxes obtained with CDTA and DTA.



FIG.5. Relative differences between group fluxes obtained with a modified ABN-set and the original ABN-set; graphite assembly, CDTA.

Spatially dependent spectra for a graphite system with an internal zone of aluminium are shown in Fig. 6. In the lower energy tail of the spectrum the flux level is almost spatially independent in the aluminium zone as a result of a large supply of moderated neutrons coming from the

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FIG. 6. Spectra in the aluminium-graphite assembly.

surrounding graphite, together with the large diffusion lengths of these neutrons in aluminium. The spectral distribution of the lower energy neutrons is strongly affected by the presence of graphite; calculations for a pure aluminium assembly give a flux envelope that strongly decreases with decreasing energy.

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### DISCUSSION

K.H. JOEST: Do you have a simple method for estimating the fitting constant B, or do you have to resort to more detailed theories with a rather fine group structure or something like the continuous slowing-down model presented in paper SM-101/55 (Cadilhac and Pujol) of these Proceedings?

H.R. KLEIJN: In our calculations we have taken B as a parameter and determined the percentage decrease in the elastic reduction crosssection. We have not yet considered simple calculations of B. However, we have performed epithermal spectrum measurements using resonance activation detectors.

### GENERATION OF INTERMEDIATE STANDARD NEUTRON SPECTRA AND THEIR APPLICATION IN FAST REACTOR PHYSICS

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#### Abstract

GENERATION OF INTERMEDIATE STANDARD NEUTRON SPECTRA AND THEIR APPLICATION IN FAST REACTOR PHYSICS. The possibility of generating intermediate standard neutron spectra through partial moderation of the uranium-235 thermal fission spectrum within a cavity in a conventional thermal column has been investigated. Multigroup transport calculations on simple spherical arrangements of source and moderator have shown that a family of well-predictable neutron spectra can be produced and that fast reactor spectra are sufficiently well simulated, in the useful energy range. A highly flexible mock-up of such a system has been realized in a spherical cavity hollowed out of the protruding end of the graphite thermal column of the BR1 reactor. Preliminary measurements by the activation technique are compared with theory. It is shown that the device can be used as a tool for better assessment of neutron spectrometry techniques and for accurate integral testing of basic nuclear data. Attention is finally drawn to a modified version of the spherical-shell transmission technique, which will tentatively be applied to improve multigroup inelastic and capture cross-section sets for some important materials.

#### 1. INTRODUCTION

1.1. The availability of standard neutron fields has long been recognized as a useful tool in the field of in-pile neutron dosimetry. For instance, the well known "standard pile" [1] [2] [3] is widely used in trying to determine absolute thermal neutron fluxes, or epithermal spectra in well moderated thermal reactors. On the high energy side, on the other hand, the  $^{235}$ U thermal fission spectrum has often been taken as a reference for spectral index measurements [4] [5]. It can easily be shown that the propagation of relative errors due to cross section shape uncertainties is greatly reduced with such a comparison procedure, while absolute errors completely cancel out.

Standard neutron spectra also are well suited for accurate integral cross section measurements, which can provide powerful checks or serve as a guide in the evaluation of microscopic differential data [6][7][8][9].

1.2. The need for non -1/E intermediate standard spectra has often been pointed out, for instance at meetings of the EURATOM DOSIMETRY WORKING GROUP. It has been put forward, in the frame of the US fast reactor physics program [10], that the intercomparison of critical experiment results, integral measurements and neutron spectra require appropriate standards.

The same question also was seriously discussed among Dutch, German and Belgian physicists and the action decided at Mol in this sense was greatly encouraged on such occasions. In the same line, J.A. Grundl from LOS ALAMOS should be especially acknowledged here for exchanges of ideas which constituted a strong support at the start of the present work.

#### 2. SCOPE OF THE INVESTIGATION

2.1. An intermediate standard neutron spectrum could be tentatively defined by the following main requirements :

- a) its shape should reasonably well simulate the spectral conditions encountered in fast assemblies
- b) its shape should therefore be variable in order to cover a broad range of hardnesses
- c) its shape should be as well known as presently possible in the energy range of interest
- d) its intensity should be sufficient for practical utilization
- e) the physical system should be simple enough to be reproducible in other laboratories.

2.2. Condition b is essential in view of the large range of neutron spectra found in fast reactors, according to the degree of core dilution. It is also evident that integral cross section measurements performed in well known spectra with highly differing mean neutron energies would be of considerable value. It consequently seems quite clear that a genuine family of intermediate standard spectra should in fact be generated, the term "family" meaning that one should be able to shift from one standard to another in a quick and easy way by changing some parameters of the physical assembly, to which the spectrum shape would be particularly sensitive.

2.3. It is imposed by condition c that the materials chosen for construction of the standard assembly have sufficiently well understood nuclear properties. Structure and canning should be reduced to such proportions as to have no practical influence on the spectra. The accuracy in the depiction of standard spectra is believed to be more important than the exactness of simulation : the energy distribution of the most important reaction rates should present the same broad features as in fast assemblies, but a detailed reproduction of spectra is in no wise considered as necessary. It will nevertheless be shown that for special purposes, more conformable "secondary" standard spectra can be generated in the same assembly, although at the expense of the accuracy.

2.4. A possible approach to the generation of intermediate standard spectra lies in the consideration of equilibrium conditions in exponential assemblies. The description of such spectra by theory would be simple and the accuracy satisfying, provided the previous requirement about constitutive materials can be fullfilled. However, flexibility required for the creation of a standard family at first seems difficult to achieve in this way; the existence of serious flux gradients and associated anisotropies, depending on the observed relaxation lengths, is not ideal for the assessment of so called "differential" neutron spectrometers, like the Li<sup>6</sup> (n,d) [11] [12] or proton recoil devices [13] [14]; finally it is not sure whether or not equilibrium could be guaranteed in practice to a sufficient degree of accuracy nor could the contribution of unwanted low energy neutrons returning to the system after interaction with the environment be predicted with any certainty.

2.5. Past experience at Mol and elsewhere has shown that it is possible to produce a pure thermal fission spectrum of  $^{235}U$  by placing a thin fissile plate inside a cavity in a moderator under such conditions that the unavoidable background neutrons can be controlled. Therefore it has been

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felt feasible to generate a family of non-1/E intermediate standard spectra by partial moderation of the 235U thermal fission spectrum in a cavity. Uncertainties about the <sup>235</sup>U fission spectrum itself will not be

discussed here. They are treated extensively in a recent LOS ALAMOS paper [16] as well as in a letter accepted for publication in Nukleonik [15].

The basic idea underlying such an approach is to choose a one-dimensional arrangement and to degrade the fission spectrum into a distribution over the whole neutron energy range from about 10 eV up to 5 MeV, by means of a material for which all pertinent nuclear data are as well known as possible. In this way, sophisticated transport calculations should provide an excellent description of the spectrum ; it is believed that the condition imposed about nuclear data restricts the choice of the material to moderators. Only H and C have been accepted at first.

2.6. In the following items, it will be discussed shortly and illustrated to what extent the five main requirements defined hereabove can be satisfied with such an approach. It will appear how the probable drawbacks noted for exponential systems are avoided.

Some experiments performed in a mock-up of the proposed standard assembly will be described and compared with theory.

Finally, possible applications in fast reactor physics will be outlined.

#### CALCULATION OF STANDARD SPECTRA AND SOURCES OF ERRORS

3.1. Design calculations have been performed by means of the 40 group GMS transport code [17] in the DS4 approximation. The GMS - II cross section library is in fact the British FD2 set [18] in the energy range of interest here. It is implicitly assumed in all the further considerations that activation foils or neutron spectrometers used in the standard spectra are covered with cadmium 1 mm in thickness. Spherical geometry was adopted.

3.2. Figure 1 displays the arrangment. It consists of a 50 cm diameter cavity embedded in a large graphite mass. The neutron source is a 1 mm thick fissile spherical shell located at the center of the cavity; this source was treated as homogeneous, but its composition was chosen to be typical of a 90 % enriched uranium oxide powder compacted by vibration into a thin stainless steel cladding.

The region labelled 2 contains an absorber whose thickness was generally fixed to  $2 \text{ gr/cm}^2$ . The role of this absorber is to prevent the low energy neutrons of the wall return spectrum to reach the inner experimental hole. The radius of the hole, noted X, is variable, depending on the thickness of region 1, which is constituted of the moderator aimed at partially degrading the source and wall return spectra. For some calculations however, other substances than moderators have also been placed in region 1.

Table I summarizes the various compositions considered for regions 1 and 2, together with the corresponding values of the intermediary radius X. Each case is characterized by a conventional number given in the first column and referred to in all subsequent discussions.

The first five cases (number 13 to 15) are relative to the so-called "polythene family of primary intermediate standard spectra"; the other ones will be discussed later on in this paper, when dealing with secondary standards or with the applications of the assembly in fast reactor physics.

13       Air $6.25$ ${}^{10}B 2 \text{ gr/ cm}^2$ 25       Polythene $5.75$ ${}^{10}B 2 \text{ gr/ cm}^2$ 21       Polythene $5.25$ ${}^{10}B 2 \text{ gr/ cm}^2$ 17       Polythene $4.25$ ${}^{10}B 2 \text{ gr/ cm}^2$ 15       Polythene $2.25$ ${}^{10}B 2 \text{ gr/ cm}^2$ 24       Mo $2.25$ $B 2 \text{ gr/ cm}^2$ 23       Fe $2.25$ $B 2 \text{ gr/ cm}^2$ 20       U 0.4 \$\$ depleted $2.25$ U 0.4 \$\$ depleted         19       U 0.4 \$\$ depleted $2.25$ B 2 gr/ cm <sup>2</sup>	erence number	Composition of region 1	Radius of internal hole X (cm)	Composition of region 2
25       Polythene $5.75$ $10_B 2 gr/cm^2$ 21       Polythene $5.25$ $10_B 2 gr/cm^2$ 17       Polythene $4.25$ $10_B 2 gr/cm^2$ 15       Polythene $2.25$ $10_B 2 gr/cm^2$ 24       Mo $2.25$ $B 2 gr/cm^2$ 23       Fe $2.25$ $B 2 gr/cm^2$ 20       U 0.4 \$ depleted $2.25$ $B 2 gr/cm^2$ 19       U 0.4 \$ depleted $2.25$ $B 2 gr/cm^2$	13	Air	6.25	<sup>10</sup> B 2 gr/cm <sup>2</sup>
21       Polythene $5,25$ ${}^{10}B 2 \text{ gr/cm}^2$ 17       Polythene $4.25$ ${}^{10}B 2 \text{ gr/cm}^2$ 15       Polythene $2.25$ ${}^{10}B 2 \text{ gr/cm}^2$ 24       Mo $2.25$ $B 2 \text{ gr/cm}^2$ 23       Fe $2.25$ $B 2 \text{ gr/cm}^2$ 20       U 0.4 \$\$ depleted $2.25$ U 0.4 \$\$ depleted         19       U 0.4 \$\$ depleted $2.25$ $B 2 \text{ gr/cm}^2$	25	Polythene	5.75	$10_{\rm B} 2 {\rm gr/cm}^2$
17       Polythene $4.25$ ${}^{10}B 2 \text{ gr/cm}^2$ 15       Polythene $2.25$ ${}^{10}B 2 \text{ gr/cm}^2$ 24       Mo $2.25$ $B 2 \text{ gr/cm}^2$ 23       Fe $2.25$ $B 2 \text{ gr/cm}^2$ 20       U 0.4 \$\nother depleted $2.25$ $U 0.4 $\nother depleted$ 19       U 0.4 \$\nother depleted $2.25$ $B 2 \text{ gr/cm}^2$	21	Polythene	5,25	<sup>10</sup> B 2 gr/cm <sup>2</sup>
15       Polythene $2.25$ ${}^{10}B 2 gr/cm^2$ 24       Mo $2.25$ $B 2 gr/cm^2$ 23       Fe $2.25$ $B 2 gr/cm^2$ 20       U 0.4 \$\$ depleted $2.25$ $U 0.4 $$ depleted$ 19       U 0.4 \$\$ depleted $2.25$ $B 2 gr/cm^2$	17	Polythene	4.25	<sup>10</sup> B 2 gr/cm <sup>2</sup>
$24$ Mo $2.25$ $B 2 gr/cm^2$ $23$ Fe $2.25$ $B 2 gr/cm^2$ $20$ $U 0.4 \%$ depleted $2.25$ $U 0.4 \%$ depleted $19$ $U 0.4 \%$ depleted $2.25$ $B 2 gr/cm^2$	15	Polythene	2.25	<sup>10</sup> B 2 gr/cm <sup>2</sup>
23     Fe     2.25     B 2 gr/cm <sup>2</sup> 20     U 0.4 % depleted     2.25     U 0.4 % depleted       19     U 0.4 % depleted     2.25     B 2 gr/cm <sup>2</sup>	24	Мо	2.25	B 2 gr/cm <sup>2</sup>
20 $U 0,4 \%$ depleted 2.25 $U 0.4 \%$ depleted 2.25 $B = 2 \ gm/cm^2$	23	Fe	2.25	$B 2 gr/cm^2$
19 II 0.4 % depleted 2.25 B.2 $gr/gr^2$	20	U 0,4 % depleted	2.25	U 0.4 % depleted
	19	U 0.4 % depleted	2.25	$B 2 gr/cm^2$
14 U 0,4 % depleted 2.25 <sup>10</sup> B 2 gr/cm <sup>2</sup>	14	U 0,4 % depleted	2.25	<sup>10</sup> B 2 gr/cm <sup>2</sup>

# TABLE I. CHARACTERISTICS OF THE VARIOUS CENTRAL SPHERICAL ARRANGEMENTS CONSIDERED IN THE CALCULATIONS

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FIG.1. Calculation model of the standard spectrum facility.



FIG.2. Typical radial flux distribution in the standard assembly.

Figure 2 shows the computed radial flux distributions through the 3.3. cavity for a few neutron groups in case 17 (table I) and illustrates the physical mechanisms leading to the production of the standard spectra ;

these phenomena are easily understood if it is noted that the wall return flux in such a cavity does not depend on position, provided there is no absorption within the source.

Figure 3 displays the standard family obtained with polythene in the conditions depicted by figure 1 and table I. On figure 4 is plotted the response function

$$I(E_{o}) = \frac{\int_{E_{o}}^{\infty} \phi(E) \sigma(E) dE}{\int_{0}^{\infty} \phi(E) \sigma(E) dE}$$

for the reaction  $U^{235}$  (n,f) in these standard spectra. It must be emphasized that no optimisation of the system has yet been carried out so that these pictures do not really represent the ultimate possibilities. The broad features of the family are nevertheless satisfactory in the sense discussed in § 2.3., the most noticeable difference with respect to actual fast reactor spectra being the absence of inelastic scattering processes so that too high a proportion of virgin fission spectrum neutrons reach the inner hole.

By introducing inelastic scattering, as is done in the secondary standard family discussed in § 6, a more exact simulation results, but at the expense of accuracy.

Graphite has also been tried as the moderator in region 1 under the same conditions : the moderating effects were too small and no family could be generated.

3.4. In all the calculations carried out so far, no effort has yet been made to determine the actual accuracy of the obtained standard spectra. This important step implies much additional work, including at least the following items :

- evaluation of the best microscopic differential cross sections and their confidence limits for H, C and B<sup>10</sup>, taking into account the most recent experimental results, including those which will be available in the near future (sec § 3.5.)
- increase in the number of energy groups until flux weighting errors become negligibly small when reducing evaluated microscopic data to the final multigroup library
- increased order of the CARLSON method and use of a sufficient number of terms in LEGENDRE polynomial angular approximations to the differential scattering cross sections (graphite mainly); it should be noted that only the first term was considered in the present investigation
- detailed study of the propagation of errors due to uncertainties on multigroup cross sections and on the representation of the uranium-235 fission spectrum.

International collaboration seems necessary to reach such objectives, which are not restricted to the scope of the present paper.

3.5. It must be emphasized that the intermediate standard spectra are in fact uranium-235 fission spectra degraded by a limited number of interaction processes of fast neutrons with selected materials : their description relies basically upon a few cross sections which have been chosen just because they are considered as primary standards [19][20] for differential energy cross section measurements.



FIG. 3. Family of intermediate standard spectra obtained via partial moderation by Polythene.



FIG.4. Response functions of the  $^{235}$  U(n, f) reaction in intermediate standard spectra (Polythene family).

3.5.1. Above 1 eV, protons contained in hydrogeneous substances can be regarded as unbound; the total cross section up to 5 MeV is practically equal to the elastic scattering cross section, which is isotropic in the centre-of-mass reference system and is known to better than 1 % [6] [19] [21] [22]

3.5.2. Below 5 MeV, elastic scattering is again the unique process to be considered in graphite  $\begin{bmatrix} 6 \\ 23 \end{bmatrix}$ , but only below 1 MeV can it be regarded as isotropic within 5 %  $\begin{bmatrix} 24 \\ 24 \end{bmatrix}$ ; between 2 and 5 MeV, the elastic scattering angular distribution varies strongly  $\begin{bmatrix} 25 \\ 25 \end{bmatrix}$  from resonance to resonance or even within one and the same resonance and doubts exist  $\begin{bmatrix} 26 \\ 23 \end{bmatrix}$ ; the angle-integrated scattering cross section is known to approximately + 5 % from 1 eV up to 5 MeV. The recent HARWELL high resolution measurements  $\begin{bmatrix} 27 \\ 27 \end{bmatrix}$  are expected to resolve the discrepancies. Some improvements might also result from refined spectrum measurements in simple geometries, as for instance the time-of-flight studies of escape spectra from graphite spheres  $\begin{bmatrix} 28 \\ 28 \end{bmatrix}$ .

3.5.3. The total cross section of B-10 is mostly composed of the (n, d)and elastic scattering contributions, the latter being effective only above 10 KeV. It seems well established [29] that the competing processes (n,t),  $(n, \chi)$  and (n,p) are practically negligible below 500 KeV, i.e. in the whole of the energy range where they could have influence for the present application. The (n, d) reaction is 1/v up to 300 keV to within 5 % [30] [31] and known to + 2 % below 10 keV and it is highly probable that the accuracy will soon reach the 1 % level in the energy range below 100 KeV. Present calculations and others [32] indicate that scattering in boron-10 has a small influence on the standard spectra ; the pertinent cross section becomes sufficiently well known [29] [31] at present for the related uncertainty to be almost negligible. The inelastic scattering effective threshold [33] lies around 4.5 MeV so that again, it is reasonable to expect no influence.

In conclusion, the significant processes involved in the generation of the proposed standard spectra are the reactions  $^{1}$ H (n,n), C (n,n) and  $^{10}$ B (n,d); the differential energy cross sections are all considered as standard; international requests with priority I and measurements under way in various laboratories will probably result in a 1% accuracy on these cross sections within a few years.

Each of these three reactions influences the standard spectra in a given energy range only so that, even though some overlapping occurs, fairly accurate theoretical description of such standard spectra should be possible.

#### 4. STATUS OF EXPERIMENTAL WORK

4.1. A mock-up of the proposed standard assembly has been constructed. Preliminary measurements have been performed by the foil activation technique, to check that no unexpected effects could hamper the practical applicability of the basic ideas developed hereabove.

It was also important to gain some technical experience about the operation of such a facility, in order to foresee any future problems and estimate the performances and to define the final configuration in all details.



FIG. 5. Exploded view of the standard spectrum facility.

As an essential part of this experimental effort, a test of wall return spectra has also been attempted, in view of their sensitivity to elastic scattering angular cross sections of graphite.

4.2. Figure 5 presents an exploded view of the facility, which is located at the external edge of BR 1 [34] horizontal graphite thermal column. The 50 cm diameter spherical cavity has been machined to a tolerance of 0,1 mm. All the new blocks used for that construction have been especially selected from the stock of pile grade B samples remaining after the erection of the thermal column itself. Great care was taken to avoid any imperfections or inhomogeneities.

The movable plug allows access to the cavity within a few minutes following reactor shut-down. Special holes normally filled with graphite have been drilled through this plug so that provision exists for introduction of air cooling tubes, electrical cables and so on. Extremely severe tolerances on the positioning and alignment of this access plug have been achieved.

The graphite thickness surrounding the cavity always exceeds 30 cm, a value which can be regarded as infinite for what concerns wall return spectra.

The central spherical shells (figure 1) can be suspended easily and accurately at the cavity centre by means of music wires. A simple tool is used for loading and unloading.

4.3. The available neutron fluxes in the thermal column and in the cavity are displayed on figure 6 for a reactor power of 1 MW. They were measured by means of gold foils 0.127 mm thick. The cadmium ratio of such foils exceeds  $10^5$  in the cavity. It has been shown that both epithermal and fast neutron leakages from the reactor core have negligible influences on the activation rates in the experiments described in § 4.4. and 4.5.,



FIG.6. Available neutron flux in BR1 graphite thermal column with the spherical cavity.

even for the less favourable capture reactions. This demonstrates that background runs (in the absence of the fission source) will probably not be necessary when operating the final standard assembly on a routine scale.

Fine angular distributions of the thermal neutron flux impinging on the central spherical shells have also been determined using copper foils. The ratio between maximum and minimum fluxes is about two and can be entirely assigned to the axial flux gradient in the bulk of the thermal column. These measurements show that it is possible to profile the thickness of the uranium shells in such a way that the fast flux in the inner experimental hole be nearly isotropic, a feature which is desirable when dealing with differential neutron spectrometers (§ 2.4.), although it is not essential for the scalar flux spectrum in that region to be identical with the spectrum expected for a uniform and isotropic thermal flux incident on a uniform source. Nevertheless, since no serious technological difficulty is associated with the realization of a suitably profiled source, calculations have been performed to establish the spectrum distortion due to neutron interactions within the source, for the nominal thickness and composition (§ 3.2.). Such distortions, though present, are found to be small and it is reasonable to hope that the future calculations will reveal a sufficiently weak dependence on the thickness in the range to be considered for complete compensation of the thermal flux gradient, so that no appreciable deviation from the one- dimensional geometry will result from the use of a profiled fission source. Since the ratio between the fission flux produced in a fissile shell and the incident thermal flux increases rapidly with shell thickness up to some maximum value, after which it de-



FIG. 7. Wall return spectrum at the centre of a spherical cavity.

creases slowly and if the optimal thickness is used on the less exposed side of the spherical shell, only a slight decrease will be necessary on the opposite side in order to produce equilibrium.

4.4. As an experimental test of the calculations of wall return spectra, radial distributions of absolute activation rates for a few foils have been measured in the cavity in a central, point fission source geometry.

Figure 7 compares the computations performed at Mol with similar DS4 calculations published by GRUNDL [35], who used the 16-group cross section library of HANSEN and ROACH [36]. Discrepancies of up to 20 % are apparent on the spectrum shapes, but the total return fluxes per unit fission source agree to better than 1 %. Thus, the comments of § 3.5.3. about graphite cross section libraries appear to be well justified.

The fission source used for the measurements was a pellet of uranium oxide enriched to 90 %, having a thickness of 2 mm, and a diameter of approximately 2 cm, cased with a thin stainless steel cladding. The activation foils all had the same diameter as the source, while their thicknesses ranged from 0.2 mm down to 0.05 mm, including dilute alloy foils supplied and assessed to 1 % accuracy by CBNM, Euratom, Geel. The foils were placed in a cadmium tube 1 mm thick and about 25 cm long, mounted on the movable access plug of the cavity, the source being fixed at the center of the opposite end-face of the tube by means of a thin aluminium ring. A special foil holder of minimum mass has been designed to guarantee an accurate knowledge of source-to-detector distances in these experiments, without introducing perturbations. It was constituted by three very thin steel ribbons graduated and held together at 90° with respect to each other by stainless steel discs located at both extremities ; the foils were slided into the graduation slots.

The activation rates all were determined absolutely by means of a  $3^{"} \times 3^{"}$  NaI (T1) crystal calibrated [37] to an accuracy better than + 2 %.



FIG. 8. Test of wall return spectrum for a point fission source at the centre of the 50 cm diameter cavity.

The results were treated as follows : the activation rate per unit fission source for the actual source-detectors geometry was computed analytically [38] in infinite vacuum for the threshold foils, as a function of the distance to the source. Similar curves were obtained for the capture reactions by normalization to the preceding case. In all cases the small perturbations within the source were accounted for ; in deriving these transport corrections, the source was simulated by a small sphere of equivalent mean chord length. Wall return contributions were added, selfshielding corrections were applied when necessary, and allowance was carefully made for the effective cadmium cut off energies in cylindrical geometry [39] . This procedure yielded the theoretical curves shown on figure 8, which are to be compared with the experimental points. The results obtained with the  $2n^{64}$  (n,p) and  $Fe^{56}$  (n,p) reactions are not included because, for the source-to-detector distances at which measurements were possible, no wall return contributions could be noticed, in agreement with theory.

Experimental errors are not expected to exceed a few percent. The theoretical curves are influenced by the choice of microscopic group cross sections for the activation detectors. An extensive evaluation work has been performed in this respect and will be published soon [40].

The theoretical wall return spectra appear to be systematically too soft, especially in the case of GMS calculations.

The discrepancy between theory and experiment is not really significant when GRUNDL's wall return spectrum is accepted. In any case, it is clear that an optimisation of the multigroup cross section library of graphite is necessary, and this could probably be achieved to a sufficient degree of accuracy during next year.

4.5. Figure 9 shows a mock-up of the central spherical arrangement, constructed to test whether true intermediate standard spectra can be produced within the cavity. The absorber is a shell of natural boron carbide with a density of 1.43 gr/cm<sup>3</sup> and a thickness of 1.7 cm, compacted by vibration within an aluminum cladding. A graphite spherical shell of 4 cm thickness is used as the moderator. The spherical source is approximated by a set of small fissile plates distributed over the outer surface of the boron screen. The complete assembly is suspended at the center of the cavity.

Figure 10 compares the central neutron spectrum in this arrangement with the core spectrum of a large dilute steam-cooled fast reactor, while figure 11 illustrates the corresponding response functions of some activation detectors.

In table II, a few measured spectral indices are compared with theory. A systematic discrepancy clearly appears, which is consistent with the observations made in the point-source geometry arrangement (§ 4.4.) and again shows the need for a better treatment of graphite nuclear data in the GMS data file. However, apart from this expected disagreement, the preliminary measurements strongly support the possibility of generating intermediate standard neutron spectra in the cavity.

Moreover, it also appears that the flux intensities which would be available in the final assembly are sufficient for most applications and that the facility would exhibit enough flexibility for the generation of a complete family of standard spectra.

#### 5. APPLICATIONS IN FAST REACTOR PHYSICS

5.1. Attention will be focused here on a few points which may be considered as the most probable objectives of our future program in connection with the facility.

The following particular applications have been selected :

- a) Reference system for spectral index measurements and for the sandwich foil technique [41] [42] [43]
- b) Reference system for checking or improving the reliability of the Li<sup>6</sup> (n, c, ) and proton recoil spectrometers
- c) Facility for accurate integral measurements of microscopic cross sections
- d) Test and improvement of non elastic and capture cross sections for structural materials through a modified version of the spherical shell transmission method [44] [45]
- e) Doppler effect measurements by the activation technique

Some short comment is presented hereafter about these five items.

5.2. Points a and b have already been referred to in the preceding paragraphs. It must be emphasized that the experimental techniques mentioned earlier are or will be used at MOL in BRO2 zero power reactor, in relation with the operation of sodium loops [46] as well as with the future SOCRATE [47] fuel testing experiment in the high flux reactor BR2. It is also worthwhile to outline that the gamma-to-neutron flux ratio in the final standard assembly will be rendered as small as a few percent, to allow proper standardization of the differential neutron spectrometers.



FIG.9. Mock-up for test of standard assembly.

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FIG. 10. Comparison of central neutron spectra in a large dilute steam-cooled fast reactor and in the mock-up of standard assembly.



FIG.11. Response functions of activation detectors in the neutron spectrum of the present mock-up.

Spectral index	Theory	Experiment
$\frac{^{63}_{Cu} (n, \chi)}{^{115}_{In} (n, n')} \frac{^{64}_{Cu}}{^{115}_{m}_{In}}$	0.64	0 <b>.57</b>
$\frac{55_{Mn} (n, 3)}{115_{In} (n, n')^{115_{m}} In}$	1.29	1,00
$\frac{197_{Au} (n, \delta)}{115_{In} (n, n')} \frac{198_{Au}}{115_{m}}$	7.75	6,50

### TABLE II. SPECTRAL INDICES IN THE MOCK-UP OF THE STANDARD SPECTRUM FACILITY

It is also planned to reduce this ratio by one decade in the existing assembly by surrounding the uranium spherical source with bismuth shells : transport calculations have shown that the resulting neutron spectrum changes are so weak that negligibly small additional uncertainties on standard spectra would result.

5.3. A systematic work is now being undertaken to redetermine the average fission spectrum cross sections for capture reactions, for it has been shown recently [15] that HUGHES' previous data  $\begin{bmatrix} 48 \end{bmatrix}$  are considerably in error; the purpose is to test  $\begin{bmatrix} 49 \end{bmatrix}$  the statistical model in the range where competition with inelastic scattering and contribution of high order neutron waves play an important role.

In the softer intermediate standard spectra, it should be possible to determine the radiation width for some individual main resonances, the other parameters being provided by differential total cross section measurements; information about strength functions could perhaps be deduced from measurements in the harder standard spectra.

Even though these integral cross section measurements cannot be interpreted directly in terms of fundamental parameters characterizing the interactions of neutrons with nuclides, no doubt exists regarding their usefulness : the general opinion is that accurate integral measurements are a necessary complement to high resolution differential studies in order to meet the overall degree of reliability on fast reactor muclear data required by considerations of safety and economics.

5.4. The spherical shell transmission method [44] [45] associated with threshold detectors has often been used in the past to give reliable information about non elastic cross sections.

Recently, serious arguments have been put forward by DAVEY [50] showing that the application of such a technique in the core of a fast





FIG. 12. Illustration of the spherical shell transmission method in the standard assembly.

critical assembly allows the unambiguous choice of the most suitable inelastic cross-section set for a given material.

The standard spectrum facility is well suited for such measurements : the improvements considered as necessary after DAVEY's exploratory research are achieved in the present system. Shells of any material with thicknesses up to 4 cm can easily be inserted inside the fission source.

The resulting perturbations have been computed in the illustrative cases of molybdenum, iron and depleted uranium with natural boron as the absorber in region 2 (figure 1 and table I, cases 24, 23 and 19). Figure 12 displays the case of molybdenum while table III gives the changes in reaction rates for a few activation detectors in the case of molybdenum and iron. A 4 cm thick depleted uranium shell results in spectrum 2 of figure 13 (see § 6) : the perturbations here are considerable in the energy range where capture is dominant, but the reaction rates are not reported because the fissions in the shell have not been taken into account so far.

For all three materials, the effects are significant at high energy: the perturbations of reaction rates or spectral indices by insertion of the shells provide a highly sensitive test of inelastic scattering cross sections. Such a test could be made even more powerful by supplementing foil activation measurements by a  $\text{Li}^6$  (n,d) spectrometer. For instance fine multigroup downscattering cross section sets could be derived, by a trial and reject procedure, over the energy range extending above the inelastic scattering thresholds. This is thought to be one of the most inte-

# TABLE III. ILLUSTRATION OF THE PERTURBATIONS TO BE EXPECTED IN THE APPLICATION OF THE SPHERICAL SHELL TRANSMISSION METHOD

	Reaction rate with shell Reaction rate without shell				
Beection					
	Shell : Mo (4 cm thick)	Shell : Fe (4 cm thick)			
<sup>238</sup> U (n,f)	0,642	0.792			
<sup>115</sup> In (n,n') <sup>115m</sup> In	0,695	0,838			
<sup>237</sup> Np (n,f)	0,862	0.950			
<sup>235</sup> U (n,f)	0.991	1.03			
<sup>63</sup> Cu (n, y) <sup>64</sup> Cu	0.793	0.989			
<sup>55</sup> Mn (n, ž) <sup>56</sup> Mn	0.605	0.936			
<sup>139</sup> La (n, 8) <sup>140</sup> La	0.794	0,963			
$197_{Au}$ (n,)) $198_{Au}$	0.706	1.03			



FIG. 13. Spectra transmitted through depleted uranium shells in the standard assembly.

resting applications of the standard facility in the field of muclear data for fast reactors.

At lower energies, using also the proton recoil spectrometer, one could hope to determine systematically in a similar way multigroup capture cross section sets, the corrections for multiple scattering in the shells being defined by comparing the normal transport calculations with similar ones neglecting scattering. Unfortunately, except in the strong low energy resonance peaks or in a few cases such as uranium-238, the shell transmission in the present arrangement is generally too high. No calculations have yet been performed to deduce the thicknesses required for such purposes. In any case, the sourcevolvity diameters would have to be increased and the cost would perhaps become prohibitive in comparison with the accuracy of the resulting information. Because the transmission is strongly affected in the predominant resonances, suitably chosen activation foils used in the same way as threshold detectors will provide useful information about some parameters of these resonances.

The most interesting application is this line probably is the study of the Doppler effect by the activation technique, heating foils or shells to increasing temperatures for various surface-to-mass ratios in both cases.

#### 6. SECONDARY STANDARD SPECTRA

Once differential spectrometers or activation foils are carefully assessed (§ 5.2.) in the intermediate standard spectra, they could in turn be used for the standardization of other, less accurately known, although reasonably reliable spectra. Such spectra are called here "secondary standards". The shell transmission technique (§ 5.4.) could greatly help in this connection.

There are two main reasons for introducing the concept of a secondary standard family :

a) improvement of fast reactor spectrum simulation by attenuation of virgin fission neutrons via inelastic scattering

b) suggestion for an assembly easier to duplicate in other laboratories.

It is felt that degrading the fission spectrum by means of natural uranium shells instead of a moderator should lead to the proper answer. Even the need for an enriched uranium shell as fissile source might be avoided in this way, depending on the applications. However, it is believed that the diameter of the cavity should be increased.

Not many calculations have yet been done in this sense. A few spectra obtained with 0.4 % depleted uranium shells are shown on figure 13 (table I, problems 20, 19 and 14; figure 1). The detailed composition of these shells is the same as SNEAK's blanket and has been selected because they are available for shell transmission experiments. The results are encouraging and indicate that, if the cavity diameter is increased, as previously suggested, boron shells could perhaps be suppressed and the thickness of natural uranium shells might then become the pertinent parameter for the flexible generation of a secondary standard family suitable in relation with modern large dilute fast reactors.

#### 7. FUTURE TREND

The performances of the standard spectrum facility could be improved by an increase of the cavity diameter. This would reduce the influence of the uncertainties in the differential angular scattering cross sections of the wall's material, probably extend the usefulness of the system to laboratories where big heavy water thermal columns are available, help generating simpler and cheaper secondary standard spectra and allow a broader application of the spherical shell transmission technique to the determination of capture and non elastic multigroup cross section sets. This would also open the way to the assessment of differential spectrometers with greater sizes as those considered here, an interesting generalization provided it is justified by some interest on the part of external laboratories.

It is not yet clear however to what extent the increased cost would become restrictive.

#### 8. CONCLUSION

It is believed that the present work strongly suggests the possibility of generating a family of non l/E-intermediate standard spectra within a spherical cavity in a conventional thermal column and sufficiently stresses the interest of such a new kind of facility for the fundamental progress of fast reactor physics.

#### ACKNOWLEDGMENTS

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Finally it must be stressed that the vibrocompacted boron shells have been perfectly realized by Mr. G. LUYTEN from BelgoNucléaire.

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#### SM-101/25

### DISCUSSION

R. VIDAL: Fission rates are measured with  $^{235}$ U,  $^{238}$ U and even  $^{239}$ Pu detectors by counting the gamma activity above a certain threshold. Can one be sure that the variations in fission product yield as a function of energy for nuclides as different as  $^{235}$ U and  $^{238}$ U are negligible? Since there is no such difficulty in the case of fission chambers, have your results been compared with fission ratios obtained by fission chamber measurements?

A. FABRY: In view of the variation in fission product yield with neutron energy, we feel that it is better not to employ activation techniques in measuring fission rates in intermediate spectra. For example, the technique based on the detection of traces in mica foils covered with a deposit of fissile material is a useful one.

J. DEBRUE: We have examined this problem in relation to  $^{235}$  U as part of the work described in paper SM-101/26. In measuring the gamma activity of fission products, a 450-860 keV window seems to eliminate the effect of the variation in fission product yield (see paragraph 3.1 of that paper), the measured activity being due principally to five isotopes with yields that vary little between the thermal energy region and 14 MeV.

TECHNIQUES DE DETERMINATION DES CARACTERISTIQUES NUCLEAIRES DE LA BOUCLE REFROIDIE AU SODIUM UTILISEE DANS BR2 POUR DES ESSAIS DE COMBUSTIBLES DE REACTEURS RAPIDES\*

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#### Abstract — Résumé

TECHNIQUES FOR DETERMINING THE NUCLEAR CHARACTERISTICS OF THE SODIUM-COOLED LOOP USED IN BR2 TO TEST FAST-REACTOR FUELS. A loop for testing fast-reactor fuels was designed and installed in the BR2 high-flux reactor. The fuel pins are cooled with sodium, which permits the removal of 130 kW of power. Absorbing screens ( $B_4$ C and Cd) round the section of the loop inside the reactor stop the low-energy components of the incident neutron spectrum. The intensity and energy distribution of the neutron flux is capable of producing a high mean power density in the test fuel without excessive flux depression, even with high fissionable-material contents. Irradiation conditions in BR2 are based on measurements made in the BRO2 reactor, the nuclear model of BR2. These values are used in conjunction with calculations made with diffusion and transport (DS<sub>n</sub> method) codes. Spectral indices and fission rates are used to check the correctness of the calculated spectrum and the constants used for the fissionable materials. The main problems consist of relating the power density in the fuel pins, as measured in BRO2, with the total power of BR2, using this power density value to calibrate the in-loop integrating flux meters, and determining changes in these parameters during irradiation. The authors discuss the problems involved in applying the combined set of measuring and calculating techniques.

TECHNIQUES DE DETERMINATION DES CARACTERISTIQUES NUCLEAIRES DE LA BOUCLE REFROIDIE AU SODIUM UTILISEE DANS BR2 POUR DES ESSAIS DE COMBUSTIBLES DE REACTEURS RAPIDES. Une boucle destinée à des essais de combustibles pour réacteurs rapides a été conçue et installée dans le réacteur à haut flux BR2. Les crayons combustibles sont refroidis par du sodium qui permet d'évacuer une puissance de 130 kW. Des écrans absorbants (B, C et Cd), entourant la section en pile de la boucle, arrêtent les composantes à basse énergie du spectre neutronique incident. L'intensité et la répartition énergétique du flux neutronique permettent d'obtenir une puissance spécifique moyenne élevée dans le combustible d'essai, sans que le creusement dans les crayons soit excessif, même pour de fortes teneurs en matière fissile. Les conditions d'irradiation dans BR2 sont déterminées à partir des mesures effectuées dans le réacteur BRO2, modèle nucléaire de BR2; ces mesures sont couplées à des calculs effectués au moyen d'un code de diffusion et d'un code de transport (méthode  $DS_n$ ). Des mesures d'indices spectraux et de taux de fission constituent un contrôle de la validité du spectre calculé et des constantes utilisées pour les matériaux fissiles. Les problèmes majeurs consistent à relier la puissance spécifique dans les crayons combustibles, mesurée dans BRO2, à la puissance totale de BR2, à étalonner par rapport à cette même puissance spécifique les moniteurs intégrateurs de flux placés dans la boucle et à définir l'évolution de ces grandeurs au cours de l'irradiation. Les problèmes posés par l'application de l'ensemble des techniques de mesure et de calcul sont discutés.

#### 1. INTRODUCTION

1.1. La boucle refroidie au sodium est l'un des dispositifs expérimentaux utilisés dans le réacteur d'essai de matériaux BR2 pour les irradiations de crayons combustibles. Cette boucle, mise en service en 1965, a

<sup>\*</sup> Travail effectué dans le cadre de l'Association CEN-Euratom chargée d'exploiter le réacteur BR2 et ses installations connexes (contrat 006/60/5 BRAB).

été conçue spécialement pour réaliser des conditions d'irradiation voisines de celles rencontrées dans un réacteur à neutrons rapides, refroidi au sodium :

- les crayons irradiés baignent dans le sodium en circulation ; la température de celui-ci peut atteindre 600 °C
- l'intensité du flux neutronique incident permet de créer, dans les crayons irradiés, une puissance linéique du même ordre de grandeur que celle caractérisant les réseaux de pile rapide
- le variation radiale de la densité de puissance dans ces crayons est minimisée grâce à l'utilisation d'écrans absorbants qui, placés autour de la section en pile de la boucle, arrêtent les neutrons de basse énergie.

1.2. La boucle et son équipement ont fait l'objet d'une description antérieure [1]. La section en pile est un doigt de gant cylindrique composé de deux chambres concentriques dans lesquelles circule le sodium ; la cavité centrale contient les crayons à irradier. Ces deux chambres sont entourées d'un écran de carbure de bore gainé ; l'enveloppe extérieure ou tube de force est en contact avec l'eau de refroidissement du réacteur.

L'ensemble est introduit dans le trou central d'un bouchon d'aluminium tapissé intérieurement d'un écran de cadmium gainé. Ce bouchon, d'un diamètre extérieur de 200 mm, occupe le centre de la matrice du réacteur.

Les pompes électromagnétiques et l'échangeur de chaleur sodium -CO, sont localisés dans une enveloppe de 200 mm de diamètre, située à l'Intérieur de la cuve du réacteur, au-dessus du coeur. Le doigt de gant et l'enveloppe forment un ensemble manipulable d'une seule pièce.

Les crayons irradiés peuvent occuper une longueur d'environ 700 mm; leur nombre peut varier de un à sept. La puissance totale évacuée est limitée à 130 kW.

La figure la est une coupe horizontale de la partie en pile de la boucle, dans le cas de l'irradiation d'un simple crayon. Les cotes sont données à titre indicatif. Des variantes de cette disposition sont réalisables, dont la plus importante consiste à supprimer l'écran de carbure de bore ; la figure lb montre une réalisation de ce type. Six éléments combustibles (type SIII) semblables à ceux utilisés habituellement dans BR2, mais d'un diamètre plus réduit (trois tubes combustibles au lieu de six) peuvent être logés dans le bouchon d'aluminium ; ces éléments sont destinés notamment à compenser la perte de réactivité qu'entraîne la présence de la boucle.

1.3. La section en pile de la boucle est exposée au flux incident des neutrons produits par les éléments combustibles (type SVI principalement) disposés autour du trou central de la matrice du réacteur. La configuration de chargement adoptée actuellement est représentée à la figure 2 ; elle est conçue de manière à répondre aux impératifs d'exploitation et aux spécifications du programme d'irradiation dans son ensemble. Une description complète des constituants du réacteur est donnée dans la référence [2].

L'intensité du flux neutronique et sa répartition énergétique sont conditionnées par les caractéristiques du flux dans la couronne de l0 éléments combustibles entourant le trou central, par la présence des constituants séparant ces éléments des écrans absorbants, et évidemment par le pouvoir absorbant de ceux-ci.

L'écran en cadmium de 2,5 mm d'épaisseur absorbe tous les neutrons en dessous de 0,5 eV environ. Il laisse passer une composante épi-



FIG. 1a. Section en pile de la boucle et du bouchon d'aluminium associé. Dispositif muni de deux écrans absorbants.



FIG. 1b. Section en pile de la boucle et du bouchon d'aluminium associé. Dispositif muni d'un écran absorbant.

TABLEAU I.	PRINCIPALES	CARACTERISTIQUES	NUCLEAIRES	RELATIVES .	A DES CONDIT	IONS TYPIQUES
D'IRRADIATIC	)N					

Type de réalisation	Caractéristiquesdu crayon irradié	Puissance spécifique maximum (W/cm <sup>3</sup> )	Creusement <u>pmoy</u> pmin pmoy (%)	Section efficace moyenne de fission <sup>235</sup> U(barns)	Energie moyenne des neutrons produisant les fissions dans U-235 (eV)	Flux d'énergie supérieure à 2 MeV (n/cm <sup>2</sup> s)
Ecrans Cd et B <sub>4</sub> C Sans éléments SIII	25 % $PuO_2 = 75 \% UO_2 (60 \% 235 U)$ Poids spécifique : 10,5 g/cm <sup>3</sup> Diamètre : 5,8 mm	1600	5	7,4	150	3,2 10 <sup>13</sup>
Ecrans Cd et B <sub>4</sub> C Sans éléments SIII	15 % PuC - 85 % UC (90 % <sup>235</sup> U) Poids spécifique : 13 g/cm <sup>3</sup> Diamètre : 10,7 mm	2000	18	5,1	350	4,4 10 <sup>13</sup>
Ecrans Cd et B <sub>4</sub> C Avec 6 éléments SIII	1d	2000	17	4,3	450	8,0 10 <sup>13</sup>
Ecran Cd Avec 6 éléments SIII	id	~ 3600	~ 30	~10	~50	8,0 10 <sup>13</sup>

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FIG.2. Configuration de chargement du réacteur.

thermique approximativement en 1/E, qui est ensuite altérée par la couche de carbure de bore de 3 mm d'épaisseur lorsque celle-ci est utilisée. L'influence des écrans et des éléments SIII sur les conditions d'irradiation d'un seul crayon est illustrée au tableau I. Ce tableau présente la valeur des caractéristiques nucléaires essentielles, pour quelques cas typiques.

La suppression de l'écran de carbure de bore augmente la puissance spécifique mais le creusement est évidemment plus important. La présence des six éléments complémentaires SIII augmente le flux au-dessus de 2 MeV<sup>1</sup> ; par contre elle laisse pratiquement inchangée la composante de basse énergie, et donc la puissance spécifique, car l'apport en neutrons pénétrant dans la boucle est compensé par l'effet de modération. due à l'eau de refroidissement de ces éléments SIII, sur le flux incident provenant des éléments SVI.

Dans les boucles de cette conception, les flux de neutrons provoquant des dégâts d'irradiation dans le gainage des crayons sont peu élevés par rapport aux puissances spécifiques réalisables dans le combustible. D'autre part, le creusement de la densité de puissance dans les crayons reste toujours relativement important. Cette situation ne peut être améliorée, dans un réacteur d'essai du type BR2, que par la création d'une zone de combustible en vue de convertir le spectre incident en un

<sup>1</sup> Le choix de ce seuil est partiellement justifié par la considération des dégâts d'irradiation dans les aciers de gainage [3], dus principalement à des neutrons de 2 MeV ou plus. Le spectre neutronique étant, dans les cas présents, identique au spectre de fission de l'uranium-235 à partir de ce seuil, toute autre valeur de flux supérieur à l'énergie E (>2 MeV) peut être calculée.

spectre voisin de celui d'un réacteur rapide. L'étude d'une boucle répondant à cet objectif fait l'objet d'une autre communication à ce symposium  $\lceil 3 \rceil$ .

1.4. Comme cela a été précisé ci-dessus, différentes variantes analogues à celles présentées à la figure 1 peuvent être réalisées, suivant les spécifications définies par l'expérimentateur.

Sur la base de ces spécifications, des calculs d'orientation sont effectués pour optimiser les conditions d'irradiation. Les paramètre considérés dans ce travail d'optimisation sont, en toute généralité :

- les dimensions des écrans absorbants, avec suppression éventuelle de l'écran de  $B_{\mu}C$
- la disposition des crayons à irradier, et dans certains cas leur diamètre et leur composition.

Le plus souvent, la teneur en plutonium des combustibles mixtes U-Pu est imposée tandis que l'enrichissement en U-235 peut être ajusté pour obtenir la puissance linéique ou la température de surface désirée.

Les calculs neutroniques sont effectués au moyen de codes de diffusion ou de transport, en géométrie cylindrique.

Les mesures préparatoires à l'irradiation sont réalisées dans le réacteur BRO2, à l'aide d'un modèle mucléaire de la section en pile de la boucle. Le réacteur BRO2 est une facilité critique dont le coeur est identique à celui du réacteur BR2 [4]; on y conduit les essais mucléaires requis par les irradiations dans BR2.

L'objectif de la campagne de mesure dans BRO2 est d'établir une prévision de la puissance spécifique qui sera produite dans les crayons fissiles (voir 3.1.). On attache toutefois beaucoup d'importance à apprécier la validité des méthodes de calcul et des constantes mucléaires utilisées, en vue d'améliorer la précision des prévisions théoriques lors des projets ultérieurs (voir 4.).

Au cours de l'irradiation dans BR2, des moniteurs de flux sont présents dans la boucle, au voisinage des crayons fissiles ; la mesure de l'activité de ces moniteurs, après leur récupération lors du démantèlement de la boucle, fournit les grandeurs de base pour l'évaluation de l'épuisement des crayons en fin d'irradiation. La calibration des moniteurs est également l'un des objectifs des mesures préparatoires à l'irradiation (voir 3.2.).

Dans les paragraphes suivants, les méthodes de calcul sont décrites ainsi que les techniques expérimentales ; les résultats de leur application à l'irradiation d'un crayon UC-PuC, en présence des deux écrans, sans éléments SIII (second cas du tableau I), sont discutés d'une manière détaillée.

#### 2. METHODES DE CALCUL

2.1. Les calculs effectués lors de l'étude d'optimisation d'une irradiation en boucle fournissent les distributions radiales du flux neutronique, en spectre et en intensité, dans le plan du point chaud. On en déduit la puissance spécifique moyenne dans les crayons ainsi que la variation de la densité de fission à l'intérieur de ceux-ci.

Vu la présence d'écrans fortement absorbants dans la boucle, il est indiqué d'employer un code de transport. Dans un grand nombre de cas cependant (voir 2.2.), l'emploi d'un code de diffusion s'avère justifié.

Les deux codes utilisés sont : le code de diffusion MODIC II [5] et le code de transport GMSII [6].

Le code MODIC II est un code de diffusion multigroupe à une dimension, écrit pour machine digitale Mercury Ferranti. Vingt-huit groupes d'énergie sont employés : 27 groupes épithermiques et un seul groupe thermique limité supérieurement à 0,683 eV. Les sections efficaces thermiques sont calculées par pondération des sections différentielles au moyen d'un spectre de Wilkins. Les constantes non thermiques proviennent de la bibliothèque GAM à 68 groupes d'énergie [7], réduite à 27 groupes selon la procédure décrite dans [8].

Le code GMSII, écrit en langage Fortran pour IBM 7030 et adapté à IBM 360, résoud l'approximation discrète S<sub>n</sub> de Carlson de l'équation de transport de Boltzmann. Le code est unidimensionnel et comporte quarante groupes d'énergie, dont huit en dessous de l'énergie 0,683 eV. La bibliothèque [9] fournit les constantes nécessaires aux calculs, sauf pour le béryllium et le cadmium. Pour ces deux éléments, des sections efficaces ont été calculées à partir des bibliothèques GAM à 68 groupes et GMSI (bibliothèque à 40 groupes dont 15 en dessous de 0,683 eV).

Tous les calculs ont été effectués en géométrie cylindrique. La boucle occupant le centre de la configuration, le réacteur est représenté par quinze à vingt régions homogènes concentriques. En ce qui concerne la boucle, le crayon fissile et chacun des écrans constituent des régions distinctes, séparées par des régions qui contiennent les différents constituants intermédiaires.

2.2. Par rapport au code de diffusion MODIC II, le code de transport GMSII a l'avantage de traiter plus correctement les écrans absorbants dans le domaine thermique où les sections efficaces de capture sont élevées et de décrire d'une manière plus détaillée le domaine thermique, chose indispensable pour le traitement des régions où le formalisme de Wilkins n'est pas applicable.

Dans les configurations à deux écrans absorbants, le flux thermique au centre de la boucle est totalement négligeable et donc également la contribution du domaine thermique à la puissance dans le crayon. Pour cette raison, un traitement théorique imparfait des écrans à forte absorption thermique ainsi qu'un calcul approximatif des constantes thermiques sont suffisants pour décrire le spectre neutronique dans la boucle. Le code MODIC II peut donc être utilisé dans ce cas (voir 4.1. à 4.3.).

En absence d'écran de carbure de bore, la contribution de la puissance dans le domaine thermique à la puissance totale n'est plus négligeable. Le code de transport GMSII à plusieurs groupes thermiques est nécessaire dans ce cas.

2.3. La figure 3 donne (traits pleins) la spectre du flux au centre de la boucle à deux écrans sans éléments SIII supplémentaires, en l'absence de crayon, calculé par code MODIC II et normalisé à  $\int_{0}^{\infty} \beta$  (u) du = l. L'ajout de six éléments SIII supplémentaires modifie la partie rapide du spectre, comme indiqué par les traits interrompus, tandis que la suppression de l'écran de B<sub>4</sub>C augmente le flux dans les domaines épithermique et thermique (traits en pointillé).

Le flux  $\int_{E}^{\phi} \phi$  (E) dE au-dessus du seuil E est également représenté. D'autres résultats théoriques sont donnés au paragraphe 4, avec les résultats expérimentaux correspondants. DEBRUE et al.



FIG. 3. Spectre des neutrons incidents au centre de la chambre d'irradiation.

#### 3. TECHNIQUES EXPERIMENTALES

Lorsque l'étude d'optimisation par calcul a fourni la meilleure solution en ce qui concerne la géométrie de la boucle et la nature des constituants, un modèle nucléaire est réalisé pour contrôler expérimentalement les principales caractéristiques du champ de rayonnement dans la cavité d'irradiation. A ce stade, les crayons fissiles ne sont généralement pas disponibles ; on mesure dans BR02 :

- la densité de fission dans le modèle, pour l'U235 et le Pu239 : f<sub>5</sub> et f<sub>9</sub>
  le flux de fission au même endroit : pour un détecteur à seuil déterminé, c'est le rapport du taux de réaction à la section d'activation pondérée sur le spectre de fission de l'uranium-235 :  $\beta_{\rm r}$  - le flux thermique sur l'axe des éléments combustibles du chargement en-
- tourant le trou central ; le flux thermique est défini par le produit de la densité neutronique en dessous de 0,5 eV par la vitesse conventionnelle de 2200 m/s : nv.

Ces grandeurs sont à la base d'une première vérification de la méthode de calcul dans les gammes d'énergie intéressant la fission et la production de dégâts dans les matériaux de gainages, par référence au flux thermique dans le chargement.

L'approvisionnement ultérieur de crayons démontables, dans lesquels le combustible est sous forme de pastilles à bases planes, permet ensuite de mesurer les densités de fission moyennes sur la section du crayon  $f_5^x$  et  $f_9^x$ .
La valeur de différents rapports de taux de réaction (indices spectraux) est enfin déterminée, avec ou sans crayons combustibles. Le creusement dans ces crayons est également mesuré.

Prévision des conditions d'irradiation dans BR2 déduite des me-3.1. sures effectuées dans BR02. Cette prévision repose sur des relations du type :

 $\frac{f_5^x}{nv} = \begin{bmatrix} f_5^x \\ f_5 \\ f_6 \end{bmatrix}_{norm} x \frac{f_5}{nv_0} x a$ (1)

Le rapport  $f_5^{x}/f_5$  caractérise la perturbation due au crayon fissile ;  $f_5^{x}$  et  $f_5$ , mesurés au cours de deux irradiations successives, sont normalisés à une même intensité du flux en un point à l'intérieur de la boucle, suffisamment éloigné du crayon dans le sens axial et donc non affecté par la perturbation locale due à celui-ci.

Le flux thermique nv est mesuré dans les éléments combustibles entourant l'expérience. Dans BR2, la valeur maximum, sur l'axe de ces éléments, est liée au flux de chaleur maximum admissible lorsque les éléments sont frais.

Le facteur a est un facteur correctif qui tient compte de ce que la normalisation adoptée plus haut pour  $f_5^{x/f}$  est arbitraire : <u>a</u> est in-férieur à l'unité de quelques pourcents.

Les valeurs des densités de fission sont déterminées par activation de disques U-Al (enrichissement 90 % U235) et Pu-Al (teneur de 97 % Pu239), à 20 % en poids de matière fissile. Des disques en uranium pur, enrichi à 90 %, sont également utilisés.

L'activité gamma des produits de fission est mesurée pour l'uranium-235, dans une bande d'énergie comprise entre 450 et 860 keV ; une série de mesures effectuées pendant la décroissance, qui suit une loi du type t<sup>-c</sup>, contribue à définir l'activité à un moment bien déterminé après l'irradiation. Pour le plutonium-239, les gamma d'énergie supérieure à 650 keV sont mesurés ; ce seuil est choisi pour minimiser l'importance relative de l'activité naturelle.

La calibration de l'installation de comptage repose sur l'irradiation des disques dans un spectre purement thermique dont l'intensité est déterminée par activation de détecteurs d'or. Le taux de comptage, dans les conditions standardisées, est ainsi mis en relation avec le taux de fission dans les disques U-Al et Pu-Al.

Pour l'uranium-235, l'influence de la variation du rendement des produits de fission avec l'énergie des neutrons  $E_n$  a été examinée ; une telle variation pourrait induire une erreur systématique en calibrant dans une colonne thermique des détecteurs utilisés dans le spectre de la boucle où l'énergie moyenne des neutrons est de l'ordre de 100 eV. Dans l'intervalle des énergies X de 450 à 860 keV, l'activité est principalement due à cinq isotopes dont le rendement ne varie pas de plus de 10 % lorsque E\_ passe de 0,025 eV à 2 MeV. D'autre part, les spectres gamma sont identiques, de même que pour un détecteur irradié dans un convertisseur ( $\vec{E}_n = 2 \text{ MeV}$ ), et les lois de décroissance ne se différencient guère. Ces arguments rendent peu probable une influence de la variation du rendement des produits de fission, pour les conditions de mesure précitées. On a pu même vérifier qu'en adoptant un seuil de discrimination inférieur de 605 keV, sans limitation supérieure, les taux de fission mesurés dans la boucle restaient inchangés. Ces conclusions conduisent finalement à affirmer qu'une variation de E de 0,025 eV à 200 eV environ peut être ignorée dans l'application de ces techniques.

Deux relations du type (1), respectivement pour l'uranium et le plutonium, fournissent une estimation de la puissance spécifique (W/cm<sup>2</sup>) moyenne à une cote quelconque du crayon considéré

$$W = \left(\frac{r_{5}^{x}}{nv_{o}} N_{5} E_{5} + \frac{r_{9}^{x}}{nv_{o}} N_{9} E_{9}\right) x nv_{o} (BR2) W/cm^{3}$$
(2)

avec N et N : nombre de noyaux d'uranium-235 et de plutonium-239 par cm

- E et E<sub>9</sub> : énergie dissipée, par fission, à l'intérieur du crayon (produits de fission, rayonnement β) : 0,272.10 Ws pour l'uranium-235 et 0,284.10<sup>-10</sup> Ws pour le plutonium-239
- nv (BR2) : flux thermique maximum, dans les éléments SVI, que l'on compte réaliser dans BR2.

La valeur de cette prévision de la puissance spécifique, sur la base des mesures effectuées dans le réacteur BRO2, doit être appréciée en tenant compte des commentaires suivants :

- 1) Les rapports  $f_{5}^{X}/nv$  et  $f_{7}^{X}/nv$  sont indépendants de toute mesure absolue car les valeurs des densités de fission de même que les valeurs du flux thermique reposent sur l'utilisation de détecteurs d'or dont l'activité est mesurée de la même manière dans les deux cas.
- 2) L'obtention d'un flux thermique maximum déterminé dans BR2 sera assurée, par un essai de courte durée, effectué à basse puissance dans BR2, au cours duquel la valeur du flux thermique maximum, mesurée par activation de détecteurs de cobalt, sera mise en correspondance avec la puissance totale du réacteur et l'indication de l'activité N16 dans l'eau de refroidissement, qui lui est proportionnelle. Cet essai est effectué au niveau de 500 kW environ ; la linéarité des instruments de mesure ont fait l'objet de tests rigoureux à plusieurs reprises, entre 500 kW et 50 MW environ.
- 3) Les éléments combustibles constituant le chargement BRO2 sont des éléments frais ; l'influence de l'épuisement qui caractérise les éléments de BR2 n'est donc pas incluse dans le résultat fourni par la relation (2). Mise à part la possiblité de simuler cet épuisement dans BRO2, on peut estimer cet effet en utilisant les informations accumulées dans BR2, relatives aux variations de  $\beta_{epi}/nv$ , où  $\beta_{epi}$  est le flux par unité de léthargie, et de  $\beta_{epi}/nv$  en fontion de l'épuisement local, et aux variations des distributions axiales de flux thermique en cours de fonctionnement [4]. La densité de fission dans la boucle est liée aux valeurs de  $\beta_{epi}$  et  $\beta_f$  dans les éléments les plus proches.

Plus simplement, on peut considérer en première approximation que, lors du fonctionnement de BR2 à puissance constante, la population des neutrons rapides est maintenue constante. La distribution radiale se modifiant peu au cours du temps, le flux rapide moyen sur la longueur active de chaque canal ne varie pas tandis que la distribution axiale s'aplatit par suite du retrait du faisceau des barres de contrôle. La densité de fission dans la boucle suit la même évolution : sa valeur moyenne, calculée sur une longueur de 700 mm environ, reste inchangée tandis que la valeur maximum décroît ; cette décroissance est obtenue par la simple considération des facteurs de forme axiaux pour les neutrons rapides, au début d'un cycle d'exploitation (barres de contrôle insérées) et à la fin du cycle (barres extraites); elle vaut environ 12 %. Ce raisonnement est confirmé par le fait que la puissance extraite de la boucle varie peu au cours d'un cycle de fonctionnement.

En conclusion, la puissance spécifique, calculée par (2) au point le plus chaud, pour un crayon fissile de composition initiale connue devrait être supérieure d'environ 6 % à la puissance spécifique moyenne au cours du cycle; ceci suppose que les éléments SVI soient frais au début du cycle.

L'expérience a montré toutefois que les prévisions basées sur les seules mesures effectuées dans le réacteur BRO2 ne peuvent être obtenues avec une précision meilleure que 10 %.

L'application des relations (1) et (2) à un crayon UC-PuC, irradié dans les conditions décrites au tableau I (second cas), a donné les résultats suivants, au point le plus chaud du crayon

> $f_{g}^{x}/nv_{o} = (4,55 \pm 0,13) \cdot 10^{-24}$  $f_{g}^{x}/nv_{o} = (5,01 \pm 0,15) \cdot 10^{-24}$

où nv<sub>o</sub> est le flux maximum mesuré dans les éléments SVI.

La composition du crayon, déjà citée, est 85 % UC - 15 % UC en poids ; l'uranium est enrichi à 90 % en U235. Le diamètre du combustible est de 10,7 mm. La puissance spécifique maximum obtenue est de 1830 + 60 W/cm<sup>3</sup>.

3.2. Détermination du taux de fission moyen dans les crayons irradiés, par mesures de l'activité des moniteurs placés dans la boucle à BR2.

Comme cela a été mentionné au paragraphe 1.4., des moniteurs sont placés dans la boucle en cours d'irradiation. Ces moniteurs sont des fils de cobalt et de fer de 9 mm de longueur et 0,5 mm de diamètre, introduits dans un microtube en acier fixé sur le tube  $\not 0$  20/23 séparant les circuits aller et retour du sodium (figure la).

Les réactions d'activation considérées sont :  ${}^{59}$ Co (n,  $\chi$ )  ${}^{60}$ Co et  ${}^{54}$ Fe (n,p) Mn54. La majeure partie de la réponse du cobalt est due à la résonance, située à 132 eV ; le fer-54 est un détecteur à seuil dont le seuil effectif est situé à 2,9 MeV. Les demi-vies sont respectivement de 5,27 années et 308 jours. Les propriétés du fer, en tant que détecteur in-tégrateur, sont satisfaisantes pour des irradiations d'une durée n'excédant pas 100 jours ; pour des irradiations de plus longue durée, d'autres moni-teurs doivent être utilisés.

Comme la réponse du cobalt est prépondérante dans une gamme d'énergie neutronique où se produisent la plupart des fissions dans l'uranium et le plutonium (voir 4.1. et figure 4), il est logique de relier l'activité des moniteurs de cobalt au taux de fission moyen, au cours de l'irradiation, dans les crayons. On minimise ainsi l'effet d'une variation éventuelle du spectre dans la boucle au cours du temps. Cette variation, qui pourrait être due à une distorsion du spectre incident en raison de l'épuisement des éléments combustibles SVI, n'a toutefois pas été décelée : la réponse du fer pourrait donc être reliée au taux de fission, de même que le cobalt. La présence des moniteurs de fer se justifie surtout pour la dosimétrie intéressant les dégâts d'irradiation.

On désigne par le symbole  $\zeta_{Co}$  le taux de désintégration par gramme d'un fil de cobalt, ayant les dimensions déjà citées, après une irradiation de l seconde. Il s'agit de déterminer les valeurs de  $f_5^X/\tau_{Co}$  et  $f_9^X/\tau_{Co}^{-}$ . Les mesures effectuées à cette fin sont résumées au tableau II.

Réacteur	BRO2	BR2	BR2		
Endroit de mesure	Modèle nucléaire de la section en pile de la boucle, avec ou sans crayons fissiles	Modèle nucléaire de la sec- tion en pile de la boucle, sans crayons fissiles	Boucle refroidie au sodium : irradiation proprement dite		
Niveau de puissance	Maximum 500 W	500 kW	55 à 60 MW		
Réactions utilisées pour les me- sures par activation	<pre>235U (n,f) 239Pu(n,f) 115In(n,n') (Eseuil = 1,1MeV) 31P (n,p) (Eseuil = 2,8MeV) 64Zn(n,p) (Eseuil = 3,2MeV)</pre>	<sup>59</sup> Co(n,Y) (E = 132 eV) <sup>54</sup> Fe(n,p) (E <sub>seuil</sub> = 2,9MeV)	<sup>59</sup> Co (n,Y) <sup>54</sup> Fe (n,p)		
Grandeurs	$ \begin{array}{ccc} f_{5}^{x}/f_{5} & f_{9}^{x}/f_{9} \\ f_{5}/\emptyset_{f}(In) & f_{9}/\emptyset_{f}(In) \\ \emptyset_{f}(In)/\emptyset_{f}(Fe)^{(a)} \end{array} $	Ø <sub>f</sub> (Fe)∕T <sub>Co</sub>	A <sub>Co</sub> (désint/s) A <sub>Fe</sub> (désint/s)		
obtenues	$\frac{f_5^{x}}{\tau_{Co}} = \frac{f_5^{x}}{f_5} \times \frac{f_5}{\varphi_f(In)} \times \frac{\varphi_f(In)}{\varphi_f(Fe)} \times \frac{\varphi_f(Fe)}{\tau_{Co}} (3)$ et, de même, $\frac{f_9^{x}}{\tau_{Co}}$		$\int f_5^{\mathbf{x}} dt = \mathbf{A}_{Co} \mathbf{x} \frac{f_5^{\mathbf{x}}}{\tau_{Co}} \int \mathcal{J}_{\mathbf{f}}^{\mathbf{f}}(Fe) dt$ $\int f_9^{\mathbf{x}} dt = \mathbf{A}_{Co} \mathbf{x} \frac{f_9^{\mathbf{x}}}{\tau_{Co}}$		
Autres gran- deurs mesu- rées	Flux thermique dans les éléments SVI	Flux thermique dans les éléments SVI Niveau de puissance	Flux thermique intégré dans les éléments SVI Variation du flux thermique dans un élément SVI au moyen d'une sonde calorimétrique à U-235		

TABLEAU II. SCHEMA DES MESURES EFFECTUEES POUR LE CONTROLE DES CONDITIONS D'IRRADIATION PAR MONITEURS DE FLUX

(a)  $p_f(Fe)$  est déduit de  $p_f(P)$  trouvé égal à  $p_f(Zn)$ ; les trois valeurs sont donc identiques.



FIG. 4. Fonctions de réponse des détecteurs utilisés pour la mesure des indices de spectre.

- Le schéma adopté résulte des considérations pratiques suivantes : - des détecteurs de fer et de cobalt, identiques aux moniteurs utilisés dans la boucle, sont impropres aux mesures dans BRO2 car leur activité est insuffisante
- les mesures dans BR2 fonctionnant à basse puissance sont effectuées au moyen du modèle nucléaire de la boucle, mais l'utilisation des crayons fissiles réservés aux mesures est exclue ; deux irradiations à cette puissance sont effectuées, dans les conditions extrêmes d'épuisement des éléments SVI.

L'application de la relation (3) (tableau II) à l'irradiation du crayon UC-PuC dont il est question en 3.1. a donné :

$$\frac{f_5^{x}}{f_5} = 0,60 \pm 0,02 \qquad ; \frac{f_5}{\rho_f(\ln)} = (30,0 \pm 1,0) \cdot 10^{-24}$$

$$\frac{\beta_{f}(In)}{\beta_{f}(Fe)} = 1,33 \pm 0,03 \qquad ; \frac{\beta_{f}(Fe)}{\zeta_{co}} = (4,97 \pm 0,08) \cdot 10^{9}$$

d'où

$$\frac{f_{5}^{x}}{f_{5}} = (1,19 \pm 0,06) \cdot 10^{-13};$$

d'autre part,  $f_{9}^{x}/f_{5}^{x} = 1,10 \pm 0,03$ ; d'où

$$\frac{f_9^x}{t_{Co}} = (1,31 \pm 0,08) \cdot 10^{-13}$$

La puissance spécifique moyenne, au cours d'une irradiation à pleine puissance de durée T, au point le plus chaud du crayon est déterminée par

$$\mathbf{w} = \mathbf{f}_5^{\mathbf{x}} \mathbf{N}_5 \mathbf{E}_5 + \mathbf{f}_9^{\mathbf{x}} \mathbf{N}_9 \mathbf{E}_9$$

$$= \left(\frac{1}{T}\right) f_{5}^{X} dt \right) \cdot N_{5} E_{5} + \left(\frac{1}{T}\right) f_{9}^{X} dt \right) \cdot N_{9} E_{9}$$

$$= \frac{A_{Co}}{T} \cdot \frac{f_{5}^{X}}{\tau_{Co}} \cdot N_{5} E_{5} + \frac{A_{Co}}{T} \cdot \frac{f_{9}^{X}}{\tau_{Co}} N_{9} E_{9}$$
(4)

La valeur maximum mesurée de A  $_{C}$  T est de 2,00.10<sup>4</sup> desint/s.g. pour une irradiation de durée unitaire ; d'où W = 1870  $\pm$  100 W/cm<sup>4</sup>.

Remarques :

- L'influence du rayonnement gamma créé dans l'ensemble du chargement de BR2 apporte une contribution supplémentaire à la puissance spécifique maximum de 100 W/cm<sup>2</sup> environ.
- 2) Les résultats expérimentaux précités ont été rendus indépendants de la perturbation du flux provoqué par les crayons fissiles, à l'endroit où se trouvent les moniteurs ; les facteurs caractérisant cette perturbation sont donnés au paragraphe 4.2.

Les techniques employées pour la mesure des flux de fission, tant dans le réacteur BRO2 que dans le réacteur BR2, ont été décrites antérieurement [10] [11] [12]. Elles reposent sur l'utilisation d'installations (spectromètre  $\chi$  dans la plupart des cas) calibrées à partir de l'irradiation des détecteurs considérés dans un convertisseur en uranium enrichi à 90 %, alimenté en neutrons thermiques par le réacteur BR1. L'intensité du spectre de fission fourni par ce convertisseur a été déduite de la mesure absolue du taux d'activation d'une série de détecteurs à seuil dont les sections efficaces sont les mieux connues.

Les détecteurs de cobalt sont mesurés par référence à des étalons, au moyen d'une chambre d'ionisation ou d'un compteur à scintillation.

4. TEST DES METHODES DE CALCUL

Les résultats expérimentaux ont été comparés aux prévisions théoriques en considérant séparément les caractéristiques suivantes :

- 1) le spectre sur l'axe du modèle nucléaire de la section en pile de la boucle, en absence de crayon combustible
- 2) les perturbations locales du flux neutronique, créées par la présence de ce crayon
- 3) la variation de la densité de puissance à l'intérieur du crayon
- 4) la puissance spécifique dans le crayon rapporté au flux thermique dans les éléments combustibles SVI constituant le chargement du réacteur.

La plupart des informations de nature expérimentale ont été obtenues par les mesures dans le réacteur BRO2. SM-101/26

4.1. Spectre neutronique sur l'axe du modèle nucléaire de la boucle. Le contrôle de la forme du spectre est basé sur la mesure d'une série de rapports de taux de réaction ou indices de spectre ; outre les réactions de fission dans l'uranium-235 et le plutonium-239, on utilise des réactions d'activation sensibles à des gammes d'énergie bien différenciées.

La figure 4 en est une illustration ; elle représente les fonctions de réponse

$$I_{1}(E_{o}) = \int_{0}^{E_{o}} \sigma_{\overline{1}}(E) \not o (E) dE / \int_{0}^{\infty} \sigma_{\overline{1}}(E) \not o (E) dE$$
(5)

des réactions utilisées, relatives au spectre calculé au moyen du code Modic II (figure 3), pour la réalisation à deux écrans, sans éléments SIII.

Le tableau III fournit les valeurs expérimentales et théoriques d'une série d'indices de spectre obtenus au moyen de ces mêmes réactions.

La calibration des installations de mesure des taux de fission et des taux d'activation pour les détecteurs résonnants est obtenue en irradiant les détecteurs dans une colonne thermique. Cette calibration repose sur la connaissance des sections efficaces thermiques ou sur des mesures absolues d'activité.

Pour les détecteurs à seuil, les mesures sont effectuées par référence au convertisseur (voir 3.2.); chaque taux de réaction est obtenu en multipliant le flux de fission mesuré par la section efficace moyenne, pondérée par un spectre de fission.

Dans la plupart des cas, les détecteurs fissiles ou résonnants se présentent sous forme d'alliage de manière à minimiser les corrections d'auto-protection.

Les valeurs théoriques, indiquées au tableau III, ont été calculées au moyen des sections efficaces ci-après :

- Dans la première colonne (A), les sections de <sup>235</sup>U et <sup>239</sup>Pu proviennent de la bibliothèque GAM [7] [8]; dans la seconde colonne (B), les mêmes sections sont basées sur la bibliothèque GMS II.
- 2) Pour les réactions  $^{197}Au$  (n,  $\chi$ ) et  $^{186}W$  (n,  $\chi$ ), on a procédé à l'évaluation des meilleurs paramètres caractérisant chaque résonance, à partir des informations les plus récentes ; ce travail d'évaluation est décrit aux références [13] et [14]. Un code [15] a été élaboré pour calculer, sous forme multigroupe, les sections et les taux de réaction à partir de ces paramètres de base.
- 3) Les sections des réactions  ${}^{59}$ Co (n,  $\chi$ ) et  ${}^{55}$ Mn (n,  $\chi$ ) sont celles de la bibliothèque GAM, sauf, en ce qui concerne le cobalt, pour le groupe contenant la résonance à 132 eV ; l'intégrale de résonance mesurée [16] a, dans ce cas, été utilisée.
- 4) Pour les réactions à seuil, les sections de groupe sont calculées en pondérant les sections différentielles par le spectre de fission de l'uranium-235 [11].

On peut déduire de la comparaison des résultats expérimentaux aux valeurs calculées les conclusions suivantes :

- Les sections efficaces tirées de la bibliothèque GMS II, pour <sup>235</sup>U et <sup>239</sup>Pu, fournissent une valeur de l'indice <sup>239</sup>Pu/<sup>235</sup>U en bien meilleur accord avec l'expérience que les sections de la bibliothèque GAM.
- 2) Mis à part l'indice <sup>186</sup>W/<sup>235</sup>U, on constate une augmentation systématique du rapport théorie/expérience lorsque l'on considère successivement les

Indice de spectre	Valeur expérimentale	Valeur théorique		Valeur théorique Valeur expérimentale	
		A <sup>a)</sup>	В	A	В
$\frac{239_{Pu}/235_{U}}{197_{Au}/235_{U}}$ $186_{W}/235_{U}$ $59_{Co}/235_{U}$ $55_{Mn}/235_{U}$ $115_{U}/235_{U}$	$1,10 \pm 0,03$ $2,12 \pm 0,03$ $1,49 \pm 0,03$ $0,35 \pm 0,02$ $0,0492 \pm 0,0012$ $(5,8 \pm 0,2),10^{-3}$	0,81 1,71 1,52 0,303 0,0462 5,84,10 <sup>-3</sup>	1,12 2,20 1,95 0,389 0,0593 7,50,10 <sup>-3</sup>	0,735 0,81 1,02 0,865 0,940 1,01	1,02 1,04 1,31 1,12 1,21 1,29
<sup>103</sup> <sub>Rh</sub> /31 <sub>P</sub> 115 <sub>In</sub> /31 <sub>P</sub>	$32,8 \pm 1,3$ 7,46 $\pm 0,23$	29,8 7,29	29,8 7,29	0,910 0,975	0,910 0,975
55 <sub>Mn</sub> /197 <sub>Au</sub> 115 <sub>In</sub> /197 <sub>Au</sub>	0,0232 <u>+</u> 0,0005 (2,72 <u>+</u> 0,05).10 <sup>-3</sup>	0,0270 3,42.10 <sup>-3</sup>	0,0270 3,42.10 <sup>-3</sup>	1,16 1,26	1,16 1,26

TABLEAU III. COMPARAISON DES VALEURS EXPERIMENTALES ET THEORIQUES DES INDICES DE SPECTRE

a) Voir texte par. 4.1.

réaction <sup>197</sup>Au, <sup>59</sup>Co, <sup>55</sup>Mn et <sup>115</sup>In, rapportées à la réaction de référence <sup>235</sup>U; ceci semble indiquer, étant donnée l'allure des fonctions de réponse, que le spectre calculé est relativement plus riche en neutrons d'énergies élevées que le spectre réel. Les valeurs des deux indices <sup>103</sup>Rh/<sup>31</sup>P et <sup>115</sup>In/<sup>31</sup>P confirment ce même effet, pour la partie supérieure à 900 keV, seuil effectif du rhodium. L'indice <sup>239</sup>Pu/<sup>235</sup>U, lui, est assez insensible à cette distorsion qui s'étend sur l'ensemble du spectre.

- 3) L'écart que présente l'indice <sup>186</sup>W/<sup>235</sup>U, dans la suite des valeurs croissantes théorie/expérience considérées ci-dessus, peut être interprété par un effet de la résonance étroite que présente le cadmium à 18,5 eV [17]; cette résonance coïncide pratiquement avec celle du tungstène à 18,8 eV. Le calcul théorique décrit probablement mal cet effet, très localisé dans le spectre, dû à l'écran de cadmium sur le flux incident.
- 4) La connaissance des sections des matériaux de détection est évidemment essentielle : les conclusions obtenues quant au spectre calculé en dépendent directement.

Les causes possibles d'une surestimation de la dureté du spectre calculé ont été examinées. La manière de décrire la configuration de chargement du réacteur, en géométrie cylindrique, dans la partie entourant immédiatement la boucle (figure 2) n'est pas sans importance à cet égard. Pour le calcul du spectre examiné dans la présente comparaison, cette partie du chargement a été assimilée à une couronne ne contenant que des cellules chargées de combustible suivie, à l'extérieur, d'une couronne représentant les cellules contenant les pièces d'extension inférieures en béryllium des barres de contrôle. On obtient un spectre moins dur, et donc plus conforme aux mesures, en homogénéisant dans une seule couronne des cellules appartenant aux deux types précités. Ainsi, les valeurs du rapport théorie/

pour 
$${}^{239}$$
Pu/ ${}^{235}$ U : 1,02  
 ${}^{115}$ In/ ${}^{235}$ U : 1,19  
 ${}^{115}$ In/ ${}^{197}$ Au : 1,12

On conserve le même accord pour le premier indice tandis que l'accord est amélioré pour les deux derniers.

#### 4.2. Perturbation due au crayon combustible

On a déterminé l'influence de la présence du crayon combustible UC-PuC sur le taux de réaction mesuré au moyen de différents détecteurs, à l'endroit où sont localisés les moniteurs de flux (paroi extérieure du tube  $\not 0$  20 - 23). Toutes les valeurs sont normalisées à une même intensité du flux en un point à l'intérieur du modèle nucléaire (ou de la boucle), suffisamment éloigné du crayon dans le sens axial et donc non perturbé par celui-ci.

Ces résultats sont nécessaires à l'application correcte de la relation (3) du tableau II (voir 3.2.) ; ils permettent d'apprécier également la validité du calcul Modic II.

TABLEAU IV. PERTURBATIONS SPECTRALES CREEES LOCALEMENT A L'INTERIEUR DE LA BOUCLE PAR UN CRAYON COMBUSTIBLE UC-PuC

Réaction	Valeur expérimentale	Valeur théorique
<sup>59</sup> Co(n,Y)	0,90	0,88
55 <sub>Mn(n,Y)</sub>	0,935	0,95
115 <sub>In(n,n')</sub>	1,15	1,20
<sup>54</sup> Fe(n,p)	1,22	1,24

Les valeurs expérimentales et théoriques sont présentées au tableau IV. Aux basses énergies, on constate une dépression du flux tandis qu'aux hautes énergies, la source neutronique que constitue le crayon provoque une augmentation du flux. Ces effets de variation sont bien décrits par le calcul théorique.

4.3. Variation de la densité de puissance à l'intérieur du crayon.

La variation radiale de la densité de puissance dans un crayon irradié est un élément déterminant de la distribution de la température. En raison de l'importance de cette caractéristique, deux techniques expérimentales basées sur l'irradiation de disques de mica placés entre les pastilles combustibles ont été mises en oeuvre. Les mesures sont effectuées dans le réacteur BRO2.

Les disques de mica, en contact direct avec le combustible, subissent l'influence des produits de fission qui créent, d'une manière très localisée, des défauts dont la densité est proportionnelle à la densité de fission  $\lceil 18 \rceil$ .

L'épaisseur des disques utilisés est de 0,045 mm. Après irradiation, ils sont plongés pendant 40 minutes dans une solution d'acide fluorhydrique qui agit comme "révélateur" des défauts et les rend propres au dépouillement ; les deux techniques de dépouillement sont les suivantes :

- a) sur des photographies obtenues au microscope avec un agrandissement x 300, on effectue le comptage des traces ; la densité des traces est d'environ  $10^{\circ}/cm^{\circ}$ ; la surface des plages examinées individuellement suivant un diamètre est de 0,07 mm<sup>2</sup>. Le parcours moyen des produits de fission dans le mica étant de l'ordre de 10  $\mu$ , les deux faces peuvent être étudiées séparément.
- b) on procède à une mesure de la densité optique au moyen d'un microphotomètre [19], d'une manière continue suivant une direction diametrale du mica ; la plage analysée a une largeur de 2 μ et une longueur de 1 mm.

Cette seconde technique a été appliquée aux micas présentant la densité de traces d'environ lo/cm<sup>2</sup>, valeur optimum pour la première technique. Une densité plus élevée est toutefois souhaitable pour les mesures au moyen du microphotomètre.



FIG. 5. Variation radiale de la densité de puissance dans un crayon combustible UC-PuC.

La figure 5 présente, réunis suivant un rayon, les résultats obtenus par analyse de trois disques irradiés simultanément, à des niveaux différents, dans l'empilement de pastilles UC-PuC. Le comptage des traces a été effectué plusieurs fois au centre et à proximité du bord pour chacun des micas, ainsi que suivant un diamètre ; en microphotométrie, trois diamètres ont été explorés pour chacun des échantillons.

C'est à la périphérie du crayon que les résultats sont le plus sujets à des erreurs systématiques. Il est probable que la distribution réelle est encadrée par les deux courbes expérimentales moyennes correspondant à chacune des deux techniques d'analyse.

On peut en conclure que le creusement calculé par voie théorique est légèrement sous-estimé. L'accord est néanmoins très satisfaisant, eu égard à la méthode de calcul utilisée.

4.4. Puissance spécifique moyenne rapportée au flux thermique dans les éléments.

Le calcul MODIC II, avec la bibliothèque des sections GAM, surestime d'une manière générale les valeurs des flux et des taux de fission à l'intérieur de la boucle. Cette surestimation est de l'ordre de 20 % dans l'exemple considéré. Cette valeur ne peut être déterminée en toute rigueur en raison de la description de la configuration de chargement par un modèle géométrique cylindrique.

C'est en raison de cette surestimation que des contrôles expérimentaux sont effectués, comme indiqué au début du paragraphe 3.

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# KINETICS (Session V)

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## ALPHA DETERMINATION FROM TRANSFER FUNCTION, OBTAINED BY PULSED NEUTRON ANALYSIS\*

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#### Abstract

ALPHA DETERMINATION FROM TRANSFER FUNCTION, OBTAINED BY PULSED NEUTRON ANALYSIS. Measurements in a sub-critical lattice have shown that the spatial dependence of the transfer functions is the same, whether they are determined by modulating a neutron source or by analysing the pulsed neutron response. When defining a transfer function for prompt neutrons, its spatial dependence can be eliminated to a high degree if only the harmonic free part of the pulse response is used for the analysis. The decay constant  $\alpha$  can be determined from these 'reduced' prompt transfer functions. The sensitivity of this method on systematic deviations from the pure exponential decay offers some valuable checks in the interpretation of pulsed neutron experiments. Also the constant background, i.e. the delayed neutron tail can be determined with high accuracy. The elimination of the spatial dependence in the transfer functions results in a correction factor for the time integral of the prompt neutron response. When it is applied to Sjöstrand's area ratio, the corrected value is in rather good agreement with the extrapolated area ratio (Gozani) and the reactivity obtained by the k $\beta/\mu$  method of Garelis-Russell.

#### 1. INTRODUCTION

In a recent paper Russell et al. [1] have shown that the (total) transfer function (T.F.) of a reactor for a neutron source can be derived from a pulse measurement, and that this transfer function agrees with the T.F. measured by source modulation. These two transfer functions show the same spatial dependence.

This paper outlines, on the basis of the work cited, how the spatial dependence of the transfer functions can be eliminated. The determination of the prompt decay constants  $\alpha$  from these harmonic free transfer functions constitutes one of the practical consequences of the method. By the same procedure the constant delayed neutron tail can be evaluated with high accuracy.

#### 2. SOURCE TRANSFER FUNCTION

A detector at a certain distance from a neutron source has a probability P(t) of detecting one emitted neutron after a time t. If the source has a (time-dependent) intensity  $S(\tau)$ , the counting rate of the detector will be

$$D(t) = \int_{-\infty}^{t} S(\tau) P(t-\tau) d\tau$$
 (1)

<sup>\*</sup> EURA TOM; contract 001-64-1 TRUB between Euratom and Association Centre d'Etude de l'Energie Nucléaire/BelgoNucléaire.



FIG. 1b. Detector counting rate for periodically pulsed neutrons.

The detection probability P(t) is the sum of two terms  $P_p(t)$  and  $P_d(t)$ , referring to prompt and delayed neutrons respectively (Fig. 1a). The integrated detector response after a single short neutron pulse ( $\delta$ -function) is

$$\int_{0}^{\infty} D(t) dt = \int_{0}^{\infty} P_{p}(\tau) d\tau + \int_{0}^{\infty} P_{d}(\tau) d\tau = A_{p} + A_{d}$$
(2)

Pulse experiments are usually performed at a constant repetition rate. If we respect the condition  $1/\alpha_1 \ll T \ll 1/\lambda_i$ , where T is the repetition period,  $\alpha_1$  is the decay constant of the fundamental mode of prompt neutrons, and  $\lambda_i$  is the decay constant of the delayed neutron group i, and if we are measuring under steady state conditions, the repetition process does not disturb the time dependence of  $P_p(t)$  (Fig.1b). Also the integral

$$\int_{0}^{\infty} \mathbf{P}_{d}(\tau) \, \mathrm{d}\tau = \mathbf{A}_{d}$$

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is retained and has now the form  $A_d = TB$ , where B is the counting rate of the constant part of the pulse response (i.e. of the delayed neutron tail). The constant detector answer B for the delayed neutrons now also makes it possible to determine the 'prompt' counting rate by a simple subtraction of B from the total counting rate.

#### 2.1. Source transfer function for prompt neutrons

Always with the condition  $T = 2\pi/\omega \ll 1/\lambda_i$ , we will now restrict our considerations to prompt neutrons only.

A sine-modulated neutron source can be represented by

$$S(\tau) = \overline{S} + \Delta S e^{j\omega\tau}$$
<sup>(3)</sup>

During the measurement of a large number of complete periods, the detector counting rate for prompt neutrons is

$$D_{p}(t) = \overline{S} \int_{0}^{\infty} P_{p}(\tau) d\tau + \Delta S e^{j\omega t} \int_{0}^{\infty} P_{p}(\tau) e^{-j\omega \tau} d\tau \qquad (4)$$
$$= \overline{D}_{p} + \Delta D_{p} e^{j\omega t}$$

By analogy with the normalized total source T.F. for a reactor [2], we now define a normalized transfer function for prompt neutrons

$$\vec{R}_{p}(\omega) = \frac{\Delta D_{p}/\vec{D}_{p}}{\Delta S/\vec{S}}$$

which is independent of the source intensity and the number of accumulated periods. Where

$$\overline{D}_{p} = \overline{S} \int_{0}^{\infty} P_{p}(\tau) d\tau = \overline{S} A_{p}$$
(4a)

and

$$\Delta D_{p} = \Delta S \int_{0}^{\infty} e^{-j\omega\tau} P_{p}(\tau) d\tau$$
$$= \Delta S \int_{0}^{\infty} P_{p}(\tau) (\cos \omega\tau - j \sin \omega\tau) d\tau \qquad (4b)$$

the real part of the prompt T.F. is

$$R_{p}(\omega) = \frac{[G_{in}^{2}(\omega) + G_{out}^{2}(\omega)]^{1/2}}{A_{p}}$$
(5)

where

$$G_{in} = \int_{0}^{\infty} P_{p}(\tau) \cos \omega \tau \, d\tau = \text{in-phase component}$$

$$G_{out} = -\int_{0}^{\infty} P_{p}(\tau) \sin \omega \tau \, d\tau = \text{out-of-phase component}$$

#### 2.2. The total source T.F. for a reactor

Considering now all neutrons, only the time-independent part of Eq.(4) must be completed. We have

$$\overline{D} = \overline{S}(A_n + A_d)$$

and the total transfer function is then

$$R(\omega) = \frac{\left[G_{in}^{2}(\omega) + G_{out}^{2}(\omega)\right]^{1/2}}{A_{p} + A_{d}} = R_{p}(\omega) M$$
(6)

where  $M = A_p/(A_p + A_d)$ . As pointed out in [2], the amplitude of the total T.F. can be related to the reactivity for an optimal frequency  $\hat{\omega}$  by

$$\rho = \frac{R(\hat{\omega})}{1 - R(\hat{\omega})}$$

The deviation of the real frequency  $\omega$  from  $\hat{\omega}$  gives only small errors for short neutron life-times. For small  $\omega$  we have indeed (Eq.(6)):  $G_{out}^2(\omega) \ll G_{in}^2(\omega)$  and  $R_p(\omega) \approx 1$ . In this case  $\rho = A_p/A_d$ , which agrees with Sjöstrand's interpretation [3].

#### 2.3. T.F. measurements

The normalized transfer function can be measured directly, point by point, by a sine-modulated source [2, 4], if one uses different frequencies for each measurement. Equation (4b) shows that it is also possible to calculate the T.F. from pulse measurements, because the time behaviour of  $P_p(t)$  is known. With  $1/\alpha_1 \ll T \ll 1/\lambda_i$  we can replace the integrals from Eqs (4a) and (4b) by sums:

$$G_{in} = \sum_{j=1}^{J_{max}} (c_j - B) \cos \omega t \Delta t$$
$$G_{out} = \sum_{j=1}^{J_{max}} (c_j - B) \sin \omega t \Delta t$$

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and

$$A_{p} = \sum_{j=1}^{j_{max}} (c_{j} - B) \Delta t$$

where  $c_j$  is the dead-time-corrected counting rate in channel number j(counts/sec),  $j_{max}$  is the total channel number, covering the whole period T, B is the delayed neutron tail of the reactor response (counts/sec),  $\Delta t$  is the channel width (sec), t is the (j - 0.5)  $\Delta t$  = time assigned to channel j (sec), and  $\omega$  is the arbitrary frequency (rad/sec).

The arbitrary frequency  $\omega$  in the weighting functions (cos  $\omega$ t) and (sin  $\omega$ t) simulates the source frequency. The range of  $\omega$  is limited for high frequencies on the channel width and on statistical fluctuation. Very low frequencies do not take the influence of delayed neutrons on the T.F. into consideration. But just the interesting part of the transfer function can be derived, and this from only one measurement. Therefore only pulse measurements will be considered in what follows.

#### 3. THE FUNCTION $P_{p}(t)$

Till now no assumption was made about the function  $P_p(t)$ . Experimental values were used for the calculation of the transfer function, found by subtraction of the constant delayed neutron tail from the total counting rates. At this stage we will assume that the time-dependent reactor response to a source pulse can be represented by a single exponential (i.e. harmonics are neglected or eliminated)

$$P_{p}(t) = X_{1} e^{-\alpha_{1} t}$$
<sup>(7)</sup>

The integrated and normalized detector response is

 $\frac{\overline{\mathbf{D}}_{\mathbf{p}}}{\overline{\mathbf{S}}} = \int_{0}^{\infty} X_{\mathbf{1}} e^{-\alpha_{\mathbf{1}}\tau} d\tau = \frac{X_{\mathbf{1}}}{\alpha_{\mathbf{1}}}$ 

Relating this to the constant delayed neutron 'background' we find Gozani's extrapolated area ratio [5]:

$$\rho_{G_0} = \frac{X_1/\alpha_1}{A_d}$$

Substituting Eq. (7) into Eqs (4a) and (4b) gives the amplitude of the prompt T.F. for an exponential response:

$$\mathbf{R}_{\mathbf{p}}(\omega) = \sqrt{\frac{1}{1 + \omega^2 / \alpha_1^2}} \tag{8}$$

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Two important consequences follow from this expression: (1) the amplitude  $X_1$  of the single exponential does not appear in the prompt T.F., and (2) because  $\alpha_1$  should not vary in space, the prompt T.F. should also be spatially invariable.

# 4. EVALUATION OF THE DECAY CONSTANT $\alpha$ FROM THE MEASUREMENTS

Because of the difference in the decay constants, the harmonics in the pulse response will generally disappear after a certain waiting time  $t_E$ . For  $t > t_E$  the remaining prompt neutron population will decay in these cases exponentially with a single decay constant  $\alpha_1$ . The time behaviour of this harmonic free part of the pulse response should be represented by Eq. (7).

Calculating the prompt T.F., Eq.(5), from the reactor response for  $t > t_E$ ;  $R_p^E(\omega)$ , we should obtain a unique T.F. at all detector locations. The shape of this T.F. should also agree with the theoretical curve. Knowing  $R_p^E(\omega)$ ,  $\alpha_1$  can be determined from Eq.(8). A suitable frequency has to be selected for this purpose. By requiring that  $\alpha_1$  be independent from the delay time  $t_E$  in the region of the exponential decay, the background due to the delayed neutron amplitude can be evaluated quite accurately. With the resulting value of the delayed neutron amplitude, it is now possible to check that the shape of the prompt transfer function for a single exponential satisfies Eq.(8) over a wide range of frequencies.

#### 5. EXPERIMENTAL INSTALLATION

The procedure described for analysing the pulse response was used for the experimental results, obtained on a highly undermoderated lattice:

(V(H <sub>2</sub> O))	1.5
moderator ratio (V(fuel))	
configuration fuel external diameter of the fuel pins fuel height in the fuel pins	cylindrical and square UO <sub>2</sub> 8.5 mm 100 cm
enrichment	1%U

The neutron source (Philips sealed tube, max.yield: $2 \times 10^9$  n/sec) was installed in the H<sub>2</sub>O reflector at midplane, 32 cm from the lattice centre.

Within the lattice, a BF<sub>3</sub> detector (5 cm active length,  $\frac{1}{4}$ -in. diameter) was used for the measurements at different locations on the x-axis (Fig. 2), and at X = -5 along the y- and z-axis. Reflector measurements were made with a 1-in. BF<sub>3</sub> detector; the monitoring was done with a BF<sub>3</sub> detector in the reflector. The detector impulses were stored in a 400 multichannel analyser (Intertechnique); the shortest measuring time was 20  $\mu$ s/channel.







FIG.3. Transfer functions in a subcritical cylindrical lattice of N = 488 fuel elements, measured at different detector positions along the x-axis at midplane.

The neutron yield of the source was adapted to the counting rate of the detector in such a manner that the dead-time correction never exceeds a few per cent.

Besides the pulse measurements, sine modulation and step variation (modified source-jerk method) were used as described in Ref. [2].



FIG.4. Prompt counting rates at different detector positions (N = 488).



FIG.5. Decay constants  $\alpha$  found by starting exponential fitting of measured counting rates at different channel numbers E.

#### 6. EXPERIMENTAL RESULTS

We will first discuss some measurements on a cylindrical lattice of N = 488 fuel elements (critical size: N = 740). The fuel-charged region of the lattice ranged from X = -12 to X = +12.

For different detector locations on the x-axis the transfer function was measured by modulating the yield of the neutron source sinusoidally.



FIG. 6. Prompt transfer functions (N = 488).

The die-away curves have been measured by pulsed neutrons under the same condition. In Fig.3 the experimental points of the (total) T.F. measured by the oscillatory neutron source are compared with the transfer functions  $R(\omega)$  obtained by analysis of the total pulse response Eq.(6). We note that both T.F. measurements exhibit the same frequency and spatial dependence.

The usual fitting of the total pulse response by a function  $f(t) = X_1 \exp(-\alpha_1 t) + B$  gives  $X_1$ ,  $\alpha_1$  and B for  $t > t_E$ . If B is subtracted from the counting rates, the time behaviour of the prompt neutron population remains. Figure 4 shows some experimental results for different detector locations. The prompt neutron decay curves were normalized at t = 1.4 msec. The shape of these curves is typical: near the source the measured points exceed the single exponential term and approach the fundamental mode only after a very long waiting time. On the other side of the reactor the counting rates rise slowly and approach the fundamental mode from below. Between both a nearly exponential decay after a short waiting time is reached: the waiting time  $t_E$  is a function of the detector location. With a convenient choice of the waiting time  $t_E$  (corresponding to a channel number E of the multiscaler) the harmonics are eliminated to a great extent.

Starting at different channel numbers E [6], the fitting gives a series of  $\alpha$ (E) from which an asymptotic value  $\alpha_{\infty}$  can generally be derived. A typical set of processed data is shown in Fig. 5. Only the measurements at X = +20 do not define an  $\alpha_{\infty}$ . All other measurements lead to asymptotic values that are, however, still slightly different from each other. The spatial dependence has thus not been eliminated completely.

Deriving the prompt transfer function from the same part  $(t > t_E)$  of the decay curve provides another check on the presence of any harmonics remaining after  $t_E$ . Figure 6 compares the prompt transfer function  $R_P^i(\omega)$ of the complete reactor response  $(0 \le t \le T)$  with the prompt transfer



FIG.7. Spatial dependence of the decay constant  $\alpha$ , the reactivity and the reactivity correction factor S (N = 488).

function  $R_p^E(\omega)$  computed from the part  $t > t_E$  of the decay curves. All transfer functions  $R_p^E(\omega)$  measured at detector locations X < +10 fall very closely together, as marked in Fig.6. The dotted line inside the marked region gives the transfer function corresponding to the characteristic decay constant of this configuration. A decay constant  $\alpha_{TF}$  is now computed with help of the prompt transfer

function  $R_p^E(\omega)$  for  $t > t_E(Eq.(8))$ . The employed frequency was  $\omega \approx 0.1 \alpha_1$  as used throughout the whole computation. Figure 7 compares some of these  $\alpha_{TF}$  with those found by fitting  $(\alpha_{\infty})$ . The two series of decay constants show nearly the same values for a wide range of detector positions X. In the neighbourhood of the neutron source, the fitting results in a rather pronounced spatial variation of the  $\alpha$ 's in the reflector. The standard deviations of the fitted values  $\alpha_{\infty}$  in the reflector (2%) do not differ significantly from those of the core measurements (1%). One could be tempted to assign equal importance to all measurements.

The transfer function can give us some more information. Because of the integration (see section 2.3) of the weighted experimental values, the influence of the statistical fluctuation on the shape of the transfer function is much smaller than in the exponential fitting. Systematic deviations from the single exponential decay can, therefore, be seen more





easily, if harmonics remain. Also, as will be explained in section 6.2, the the prompt part of the decay curve is treated separately from the constant part. An incorrectly estimated background amplitude B cannot, therefore, be compensated by a variation in the exponential term as is the case in the fitting procedure.

Figure 8 compares the  $\alpha_{TF}$  from the measurements at X = -9 and X = +15. Only the measurement at X = -9 gives a constant  $\alpha_{TF}$  for a large range of waiting times  $t_E$ (corresponding to a channel number E). At X = +15 the measuring time was much too small; the influence of harmonics persists for such a long time that prompt neutrons of the fundamental mode are completely swamped.

Figure 9 leads to the same conclusion. At X = +15 no starting channel number E can be found for which the decay constant is independent of the circular frequency. At X = -9 and E = 21 the value of  $\alpha_{\rm TF}$  remains constant until high frequencies which indicates an exponential decay. The fluctuation of the measured values becomes visible at very high frequencies only, but the decay constants start oscillating at increasingly low frequencies if the delay time t<sub>F</sub> is enlarged.

The presence of harmonics is only one reason for the dependence of  $\alpha_{\rm TF}$  on the starting channel number E. When determining the delayed neutron tail, the residual error also results in a dependence of  $\alpha_{\rm TF}$  on E. This property can be used to compute the amplitude of the time-independent part of the detector response.

Figure 10 demonstrates the sensitivity of this method. The measurement of the pulse response at X = -5 in a cylindrical configuration of N = 204 fuel elements was fitted by an exponential. A first estimate  $B_{\Sigma}$  of the constant delayed neutron background was found by averaging the counting rates over the last fifty channels (total channel number used in this measurement: 200). By the fitting, another background  $B_{\infty}$  was found. Using these two values for computing the transfer functions, the values derived for the decay constants vary with the starting channel number E. By requiring  $\alpha$  to be constant over twenty channel widths (between E = 25 and E = 45), another estimate of the background  $B_{TF}$  was found. This method was used for all examples of decay constants given here when they were determined via transfer functions.



FIG.9. Variation of decay constants  $\alpha$  using different circular frequencies in the prompt transfer functions.



FIG.10. Dependence of decay constants  $\alpha_{\rm TF}$  on the subtracted constant amplitude B.



FIG. 11. Prompt transfer functions in a reflected square configuration of  $N \stackrel{\circ}{=} 25^2$  fuel elements.



FIG. 12. Prompt transfer functions in a square lattice of  $N \triangleq 25^2$  fuel elements with highly poisoned reflector.

The methods described here have also been applied to measurements made in a square configuration of N =  $624 \, \stackrel{\circ}{=}\, 25^2$  fuel elements. In one case the lattice was normally reflected (H<sub>2</sub>O); in the other the reflector was highly poisoned with boron (~ 5500 ppm B). Figures 11 and 12 give the transfer functions  $R_p^i(\omega)$  and  $R_p^E(\omega)$  for the reflected and the poisoned assembly respectively. The decay constants computed from the  $R_p^E(\omega)$  are displayed in Fig.13.

There is still another check on the validity of our procedure for eliminating harmonics in the transfer functions. Until now our attention has been focused on the frequency behaviour and related properties of the



FIG.13. Decay constants  $\alpha_{TF}$  in the square configuration N  $\stackrel{\circ}{=} 25^2$  with poisoned and unpoisoned reflector.



FIG. 14. Distribution of the 'prompt' area  $A_p$  and the 'delayed' area  $A_d$  in the cylindrical configuration of N = 488 fuel elements.

normalized prompt transfer function. The consequences on reactivity determination are also worth investigating.

The ratio of the time integrals of the prompt to delayed responses gives the Sjöstrand expression for the reactivity  $\rho_{sj} = A_p/A_d$ . This was related in section 2.2 to the amplitude of the total transfer function if a convenient circular frequency was used:

$$\rho_{sj} \approx \rho = \frac{R^{i}(\hat{\omega})}{1 - R^{i}(\hat{\omega})}; \text{ with } R^{i}(\hat{\omega}) = R^{i}_{p}(\hat{\omega}) \frac{A_{p}}{A_{p} + A_{d}}$$



FIG. 15. Spatial dependence of reactivities and decay constants along the z-axis in the reflected square configuration of N  $\stackrel{\circ}{=} 25^2$  fuel elements.

As shown in Fig. 7, the values of  $\rho_{sj}$  have a very marked space dependence which is due to the presence of harmonics. Figure 14 illustrates the well-known fact that these harmonics are mainly present in the prompt area. To reduce the influence of these harmonics the prompt area has to be adjusted. We look for a correction factor S to convert this prompt area, which contains all harmonics

$$(A_{p} = \sum_{i}^{n} x_{i} \alpha_{i}, i = 1, 2, 3 \dots n)$$

into the prompt area of the fundamental mode  $X_1/\alpha_1$ . S can thus be interpreted by  $S = (X_1/\alpha_1)/(\sum_{i=1}^{X_1/\alpha_1})$  and we get a corrected reactivity

$$\frac{SA_{p}}{A_{d}} \approx \rho^{*} = \frac{R^{E}(\hat{\omega})}{1 - R^{E}(\hat{\omega})}, \text{ with } R^{E}(\hat{\omega}) = R_{p}^{E}(\hat{\omega})\frac{SA_{p}}{SA_{p} + A_{d}}$$

The definition of S is given by

$$\frac{\rho^*}{\rho} = S = \frac{R_p^{E}(\hat{\omega})}{R_p^{i}(\hat{\omega})} + \frac{1 - R_p^{i}(\hat{\omega})}{1 - R_p^{E}(\hat{\omega})} + \frac{A_d}{A_p} + \frac{1}{R_p^{i}(\hat{\omega})} \left(\frac{1 - R_p^{i}(\hat{\omega})}{1 - R_p^{E}(\hat{\omega})} - 1\right)$$

The important term for the elimination of harmonics is given by

$$\frac{1 - R_p^1(\hat{\omega})}{1 - R_p^E(\hat{\omega})}$$

Computing the S values for each measurement in the cylindrical configuration of N = 488, we obtain the results presented in Fig. 7. Multiplying the area ratios of Sjöstrand with S, we get the corrected reactivities  $\rho^* = S \rho_{si}$ . They are nearly constant over a large part of the reactor core

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with exception of the regions in the vicinity of the boundaries. Furthermore they agree quite well with the reactivities calculated by the  $k\beta/\ell$  method of Garelis and Russell [7]. This lends some evidence to the conjecture that both methods eliminate the harmonics to the same extent.

Another interesting example is displayed in Fig.15. It was obtained in the square configuration  $N \stackrel{\circ}{=} 25^2$  (with H<sub>2</sub>O reflector). The measurements were made at X = -5 and at different distances of Z from the midplane. We note that  $\rho_{sj} = S \rho_{sj}$ , i.e. S = 1, in the region where the node of the third harmonic can be expected.

#### 7. CONCLUSION

It has been illustrated experimentally that the spatial dependence of the transfer functions is related to the transient behaviour of the prompt neutrons immediately after a neutron pulse. Using only the exponential part of the decay curve, a nearly spatial independent transfer function for prompt neutrons can be derived. The decay constants  $\alpha$  can be computed from this 'reduced' transfer function.

The high sensitivity of the prompt transfer function on systematic deviations from the exponential law offers a valuable check in the interpretation of the decay curve of the prompt neutrons, which is of great help in the comparison of  $\alpha$  measurements made at different detector locations.

In addition, the constant part of the reactor response can be determined with high accuracy. This leads to a higher precision in reactivity measurements, especially in the presence of a high natural background, as is the case with plutonium-fuelled assemblies.

The elimination of the spatial dependence in the transfer function still has another consequence: knowing the amplitudes of the transfer functions with and without harmonics, a correction factor can be found which removes the discrepancy between the reactivities obtained by using the area ratio (Sjöstrand) on the one hand, and the extrapolated area ratio (Gozani) or the  $k\beta/\ell$  method (Garelis and Russell) on the other hand.

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#### DISCUSSION

F. EBERSOLDT: You have assumed an exponential decay for the neutron population to be detected, though I believe that the regions studied were highly sub-critical. If this is the case, it no longer seems justifiable to assume single-mode decay. Have you calculated or estimated the error in the detected neutron balance introduced by suppression of the other modes? What were the  $k_{eff}$  values of the regions investigated?

W. ROTTER: The sub-critical value of the lattice was 9.3 \$. The method has also been applied to a lattice of 204 fuel rods (with a sub-critically of ~25\$) with the same success, since we did not 'assume' exponential decay; only that part of the decay curve (after a certain time-lag  $t_E$ ) where the curve appears to be exponential was used. The 'reduced' transfer function  $R_E^F(\omega)$  then indicates whether the decay curve is in fact a pure exponential or not.

## TIME-DEPENDENT S<sub>N</sub> CALCULATIONS DESCRIBING PULSED SOURCE EXPERIMENTS AT THE FRO REACTOR

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#### Abstract

Time-dependent  $\boldsymbol{s}_N$  calculations describing pulsed source experiments at the fro REACTOR. In view of the difficulties in describing pulsed source experiments quantitatively in assemblies consisting of a fast core and a light reflector, a time-dependent  $S_N$  code has been applied to this type of assembly. The code, written for the IBM 7090 computer, divides time into short intervals and computes the flux in spherical geometry for each interval using the Carlson  $S_4$  scheme. The source term is obtained by extrapolation from two earlier time-intervals. Several problems in connection with the discretization of the time, space and energy dimensions are discussed. For the sub-critical assembly studied the treatment of the lower energy-groups is decisive for the numerical stability. A 22-group cross-section set with a low energy cut-off at 0.04 eV obtained with the SPENG programme has been used. The time intervals are varied continuously and are set proportional to the inverse of the maximum logarithmic time-derivative of the space and energy-dependent flux with the further restriction that they are not allowed to increase above a predetermined value. In a typical case, the intervals vary between  $10^{-9}$  and  $10^{-8}$  sec. The memory of the computer is fully exploited when 22 energy groups and 46 radial points are used. The computing time for each time-interval is about 6 sec. The code has been applied to a 3.5% sub-critical assembly consisting of a 20% enriched, spherical uranium metal core with a thick copper reflector and the calculations have been compared to experiments with good agreement. The calculations show that spectral equilibrium below 10 keV is not reached until times long compared to the usual measuring times and that the exponential decay finally reached is entirely determined by reflector properties at almost thermal energies. It is also shown that the simple one- and two-region models are inadequate in this case and that no time-independent prompt neutron life-time can be obtained from the measurements.

#### 1. Introduction

Pulsed source experiments in multiplying systems have usually been analyzed with the point reactor model. In this model it is assumed that spectral equilibrium is attained rapidly throughout the reactor and that the measured decay of the neutron population is given by the same exponential (except for rapidly decaying higher modes) independently of the spectral sensitivity and the position of the detector used. The decay constant is in this model a simple function of the reactivity of the system, the effective delayed neutron fraction and the mean prompt neutron lifetime which are all easily defined parameters.

Brunson, Huber [1] and Waltar, Ruby [2], respectively, have reported experiments on heavily reflected systems characterized by very unequal neutron life-times in core and reflector, taken separately. In these cases, the simple point reactor model has turned out to be inadequate, but the "two-point model" proposed by Cohn [3] seems to be able to explain the experimental results. In this model, core and reflector neutron populations are treated separately. Besides the core multiplication constant, interaction parameters between core and reflector populations and separate neutron life-times have to be defined. These parameters are supposed to be time independent. The model predicts a prompt neutron decay characterized

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by two exponentials with the same decay constants throughout the reactor, but with different relative amplitudes in core and reflector. The fast decaying exponential is mainly given by reflector parameters, while the slower one corresponds to the main mode of the reactor and is reactivity dependent.

Neither of the models seems to be applicable to pulsed source experiments performed at assembly 2 of the FRO reactor. This assembly consists of an undiluted 20 % enriched uranium core and a thick copper reflector. Due to the low absorption and rather good moderating power in copper together with the low leakage rate out of the reflector, an appreciable number of the fast neutrons coming from the core has time to reach very low velocities before disappearing from the system. This means that the characteristic decay constant of the reflector is very low and it turns out that even in the critical reactor this constant is smaller than that determined by the multiplication in the core. Since there is no upscattering of neutrons in the reflector this also means that the reflector spectrum is changing during the whole measurement and consequently also the probability that a reflector neutron returns to the core. This makes one of the assumptions in the two-point model invalid.

#### 2. Numerical solution of the Boltzmann equation

In multigroup formalism, the time-dependent one-dimensional Boltzmann transport equation will read

(2.1)

$$\frac{1}{v_{k}} \frac{\partial}{\partial t} N_{k}(t,r,\mu) + \mu \frac{\partial}{\partial r} N_{k}(t,r,\mu) + \frac{1-\mu^{2}}{r} \frac{\partial}{\partial \mu} N_{k}(t,r,\mu) +$$
$$+ \Sigma_{n} N_{n}(t,r,\mu) = S_{n}(t,r,\mu) \dots$$

where

v <sub>k</sub>	=	neutron velocity in group k	
r	=	space coordinate	
ų	=	cosine of angle between flux and radius vector	
N <sub>k</sub> (t,r,µ)	=	angular flux in group k	
Σk	=	transport cross section	
S <sub>k</sub> (t,r,µ)	=	source term	

The source term can be expressed as a sum  $S_k = S_k^i + B_k$  where  $S_k^i$  originates from fissions and scattering and  $B_k$  comes from external sources.

For the numerical solution of eq. (2.1) we have used a code employing the Carlson  $S_N$ -method. This code has been described earlier (4) and we will in this paper only discuss the discretisations made in the rtEµspace.

#### 2.1 Energy grid

Several attempts were made with different energy grids in order to establish a valid discretisation of the energy dimension. This investigation proved to be extremely important since it turned out that a 14-group

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cross section set was insufficient for the case we had with considerable slowing-down occurring in the reflector. For a 14-group case we even obtained a divergence of the computational process which could not be remedied by changing time or radial discretisation and which occurred after a practically constant number of time-steps regardless of their length. We ascribe this phenomenon to a feed-back via slow fission to the higher energy-groups of errors in the lower groups caused by an improper discretisation there. When using a 22 group cross section set these difficulties disappeared. Different values of (1/v) were used in core and reflector but this proved to give an almost negligible improvement compared to the case when they were set equal in core and reflector.

#### 2.2 Radial grid

Also here several grids were tried in order to establish a valid discretisation. The choice here turned out to be less critical as long as the grid was made finer close to the core-reflector boundary.

#### 2.3 Time grid

For the discretisation of the time dimension we used a method similar to that described in [5], and varied the time-steps continuously setting them inversely proportional to the largest absolute value of the logarithmic time-derivative of the flux in space and energy, and with the further restriction that the time-step was not allowed to increase above a predetermined value of the order of  $10^{-0}$  sec.

Several computational schemes were then investigated in an attempt to optimize the number of time-steps required. The following three methods were compared in this respect.

- 1) In the method 1, the one previously used in ref. [4], the source term  $S_k$  at time t 0.5 $\Delta$ t is determined by extrapolation. In the term  $\Sigma_k N_k$ , we set  $N_k = 0.5(A+B)$ . Here A is the flux at time t- $\Delta$ t, which is computed in the earlier step and B is the flux at time t.  $S_k^i$  is determined as  $S_k^i = S + 0.5(S-S^1)$ , where S is the source term at time t- $\Delta$ t, and  $S^1$  is the source term at time t-2 $\Delta$ t.
- 2) In method 2 we set for  $N_k$  and  $S_k^i$

 $N_k = N_k(t-\Delta t)$ , i.e. the flux at time  $t-\Delta t$  $S_k = S(t-\Delta t)$ , i.e. the source term at time  $t-\Delta t$ .

3) In method 3, a one-step iteration is performed, where the values of  $N_{\nu}(t)$  and S(t) from method 2 are used again in the same time-step.

$$N_{k} = 0.5(N_{k}(t-\Delta t) + N_{k}(t))$$
  

$$S_{k}^{i} = 0.5(S(t-\Delta t) + S(t))$$

Contrary to what might have been expected since they contain no extrapolations, the stability of methods 2 and 3 was decreased compared to method 1, so that a shorter time-step was required. If a shorter time-step was used, however, there is very good agreement between the results obtained from methods 1 and 3 and acceptable agreement with the results from method 2. If  $\Delta t$  is chosen 0.2.10<sup>-8</sup> sec in our problem, all three methods will be stable. The methods 2 and 3 show comparable accuracy. In method 1 we could use a time-step  $\Delta t = 10^{-8} - 1.5 \cdot 10^{-8}$  sec. Since method 3 requires twice the computation time of method 1 for each time-step, method 1 will be superior. We will now study why method 1 shows greater stability than the others.

We then analyze the stability behaviour of one of the group equations under some simplified assumptions.

- i) We assume the source term  $S_k^i$  not to be critical for the stability since it consists of a sum of integrals.
- ii) We assume that for stability considerations we can neglect all derivatives except the time-derivative.

Under these assumptions we obtain the following equation for a perturbation y

$$\frac{1}{\mathbf{v}_{k}} \quad \frac{\mathrm{d}\mathbf{y}}{\mathrm{d}\mathbf{t}} = -\Sigma_{k}\mathbf{y}$$

For method 2 this gives

$$\mathbf{y}(t) = \mathbf{y}(t - \Delta t) - \Delta t \mathbf{v}_{1} \boldsymbol{\Sigma}_{1} \mathbf{y}(t - \Delta t) = (1 - \Delta t \mathbf{v}_{1} \boldsymbol{\Sigma}_{1}) \mathbf{y}(t - \Delta t) .$$

If  $\Delta t \in \Sigma$ , > 2 the perturbation will grow from step to step and we will have an instability.

Method 2 was unstable for  $\Delta t = 0.5 \cdot 10^{-8}$ , which we also can deduce from the above argument. For energy group 1 we have  $v = 24 \cdot 10^8$  cm/sec and  $\Sigma = 0.12$  cm<sup>-1</sup>, which makes  $\Delta t v_{\perp} \Sigma_{\perp} = 2.06$ .

For method 1 we have  $y(t) (1+0.5\Delta tv_k \Sigma_k) = (1-0.5\Delta tv_k \Sigma_k)y(t-\Delta t)$ ,

i.e. 
$$y(t) = \frac{1 - 0.5 \Delta t v_k \Sigma_k}{1 + 0.5 \Delta t v_k \Sigma_k} y(t - \Delta t)$$
.

This factor is < 1 for all  $\Delta t$  so the type of instability discussed above cannot occur.

Method 3 gives  $y(t) = y(t-\Delta t) - 0.5 \Delta t v_k \Sigma_k (y(t-\Delta t) + (1 - \Delta t v_k \Sigma_k) y(t-\Delta t))$   $y(t) = y(t-\Delta t) (1-\Delta t v_k \Sigma_k + 0.5 (\Delta t v_k \Sigma_k)^2)$ This becomes unstable for  $\Delta t v_k \Sigma_k > 2$ .

#### 2.4 Angular grid

When the discretisation of the r, t, E dimensions was performed in accordance with the description above, the agreement between  $S_L$ -calcula-

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tions and experiment turned out to be satisfactory as we shall see later. For this reason and computation time considerations no investigation was made of variations in the angular discretisation.

## 3. Cross section set

The group cross sections used were obtained by the SPENG [6] program from cross section tapes currently in use at AB Atomenergi. The cross sections are in the tapes given pointwise, except in the resonance regions, where narrow group cross sections have been calculated using resonance parameters. When the calculations were made only an old set of copper cross sections given pointwise was available. A more modern set gives appreciably lower absorption cross section in some groups below 5 keV. For each region the SPENG program calculates a weighting fine structure spectrum using fundamental mode B1-approximation taking shielding of resonances into account. Since it turns out that very low energy neutrons are of importance, a 22 group set with a lethargy step of 0.9 in most groups and a low energy cut-off at 0.04 eV was constructed (cf. Table I). No upscattering was included. Since the calculations are to deal with prompt neutrons only, the fractions of fission neutrons in each group were multiplied by  $(1 - \beta)$ . Besides the cross sections entering steady state calculations, 1/v and detector cross sections were averaged over the appropriate spectra.

### 4. Experiment

Since the experimental set-up has been described in [4, 7] only some aspects of it will be discussed here. The most thoroughly investigated assembly (Assy 2) consisted of an approximately spherical undiluted 20 % enriched U<sup>235</sup> core surrounded by a cylindrical copper reflector, more than 35 cm thick. The assembly was made 3.5 % subcritical by a reduction of the core radius. In order to keep the spherical symmetry the tritium target, producing bursts of 14 MeV neutrons, was introduced to the core center through a 2.7 cm square channel. As neutron detectors we used a He<sup>3</sup> filled gas scintillator and a hydrogen filled spherical proportional counter. In both counters it was possible to cut off the gamma pulses with a simple discriminator. The spectral sensitivity of these detectors is given in Table I. When the gas scintillator was used the time resolution of the electronic set-up was given by the channel width of the multichannel analyzer in the actual run (0.2 or  $0.4 \ \mu s$ ), while the time resolution of the proportional counter was about 2 µs. Three detector positions were used, one in the core, one at the core-reflector boundary and one in the reflector. The burst repetition rate was varied so as to keep the ratio between peak intensity and delayed neutron background at about 100.

## 5. Comparison between theoretical and experimental results

The results of experiments and calculations on the subcritical Assy 2 are given in Figs. 1-3. The calculations were made on a spherical core surrounded by a 35 cm thick spherical reflector. In order to take into account the reactivity effects of target and detector channels, the critical radii with and without channels were determined and the experimental subcritical radius was decreased in the same proportion (to 16.28 cm) in the calculations. In the figures shown, the experimental curves have been normalized to the pertinent theoretical ones at 8  $\mu$ s. Due to an uncertainty in the experimental zero time the curves have been shifted in the time

<u> </u>							
Group	Energy boundary (MeV)	He <sup>3</sup>	H <sub>2</sub>	Group	Energy boundary (MeV)	He <sup>3</sup>	H <sub>2</sub>
	10.0						
1	2.25	2.04	2.09	12	$8.8 \times 10^{-4}$	21.5	-
2	1.25	1.74	3.05	13	$3.5 \times 10^{-4}$	34.9	-
3	7.0 $\times 10^{-1}$	0.847	3.98	14	$1.4 \times 10^{-4}$	57.0	
4	$3.9 \times 10^{-1}$	0.826	4.83	15	$5.5 \times 10^{-5}$	88.2	
· 5	$2.2 \times 10^{-1}$	1.039	5.15	16	$2.2 \times 10^{-5}$	146	-
6	1.2 × 10 <sup>-1</sup>	1.48	3.86	17	$8.8 \times 10^{-6}$	217	-
7	$6.1 \times 10^{-2}$	2.16	0.37	18	$3.5 \times 10^{-6}$	308	· _
8	$3.4 \times 10^{-2}$	3.24	-	19	$1.4 \times 10^{-6}$	505	-
9	$1.5 \times 10^{-2}$	4.84	-	20	$5.5 \times 10^{-7}$	796	-
10	$5.5 \times 10^{-3}$	7.52	-	21	$2.2 \times 10^{-7}$	1230	-
11	$2.2 \times 10^{-3}$	13.3	-	22	$4.0 \times 10^{-8}$	. 1940	-
	<u> </u>						

## TABLE I. SPECTRAL EFFICIENCY OF DETECTORS



FIG.1. Decay of the neutron population measured and calculated for a detector position (r = 10.4 cm) in the core of Assy 2. The slope of the straight lines is obtained by stationary  $S_4$  calculations.



FIG.2. Decay of the neutron population measured and calculated for a detector position (r = 23.5 cm) at the core reflector boundary of Assy 2. The slope of the straight lines is obtained by stationary  $S_4$  calculations.



FIG.3. Decay of the neutron population measured and calculated for a detector position (r = 44.5 cm) in the reflector of Assy 2. The slope of the straight lines is obtained by stationary S<sub>4</sub> calculations.



FIG.4. Decay of the neutron population measured in the reflector of Assy 2 with an He<sup>3</sup> detector at addifferent reactivities (from top: 0.69, 1.29 and 3.59% below critical, respectively).





## direction to reproduce the peak intensity (when discernible) or to give the best overall fit.

As is seen the agreement between experiment and time dependent  $S_4$ calculations is good. The discrepancy in the slope of the final decay (13 \$) for the He<sup>3</sup> detector in the reflector position (Fig. 3) might be explained by the fact that the absorption cross section in copper ought to be decreased and that the calculations were made with a spherical reflector while the experimental reflector was cylindrical and somewhat larger. The slope of the straight lines drawn in the figures is obtained by a stationary  $S_4$  calculation, where an exponentially decaying flux was postulated and the slope determined by iteration. In these calculations only 2 instead of 8 groups were used below 100 eV.

## 6. Discussion

The good agreement between experiment and theory shown in Figs. 1-3 justifies confidence in the results of the time-dependent code even in time regions where no detailed experimental results are available - at least until new phenomena such as thermal up-scattering start to become of importance. Since the model used in the calculations with a group structure in energy and space and a finite low energy cut off implies the existence of a discrete set of eigenvalues to the Boltzmann equation it is of interest to investigate which part of the reactor and which energy group is responsible for the slowest decaying mode calculated and if this mode has a physical significance.

The experimental results shown in Fig. 4 indicate that the final decay is entirely given by the properties of the reflector. These measure-



FIG.6. Removal rates in low-energy groups of the copper reflector.

ments were made with the He<sup>3</sup> detector in the reflector of Assy 2 at three different reactivities. It is seen that the same exponential is reached in all cases.

Fig. 5 is an illustration of the variation in neutron spectrum during the decay. It gives, besides the calculated response of the He<sup>3</sup> detector in a reflector position, the neutron densities in different low energy groups as a function of time. Apparently the total neutron density which is measured by the He<sup>3</sup>-detector in low energy groups - decays exponentially although the neutron spectrum changes with time. A more careful investigation of the calculations shows that the decay constant decreases 3 per cent between 25 and 45 µs and 0.7 per cent between 45 and 80 µs after the pulse injection - just a little more than the rounding-off errors introduced when the results are printed by the computer. This approximately exponential decay could be explained if the sum of the leakage and absorption rate were independent of neutron energy. In diffusion approximation this would be the case if

 $\alpha = \mathbf{v}(\mathbf{D}\mathbf{B}^2 + \boldsymbol{\Sigma}_{\alpha})$ 

(6.1)

were energy independent. Below the first positive resonance at 229 eV the absorption cross section for copper ( $\Sigma_c$ ) approaches 1/v for decreasing energies, but due to a resonance in Cu<sup>63</sup> at - 100 eV it lies below this value down to about 5 eV. Since the diffusion constant (D) does not change much with energy it might be possible to find a buckling (B<sup>2</sup>) that makes expression 6.1 constant in a finite energy region. Fig. 6 gives v $\Sigma_c$  calculated in the low energy groups for the actual reflector composition. The decay constant for the neutron density from the time dependent S<sub>4</sub> calculations ( $\alpha = 7.4 \cdot 10^4 \text{ s}^{-1}$ ),  $v(DB^2 + \Sigma_c)$  and  $v(DB^2 + \Sigma_{rem})$  are also shown.



FIG. 7. Calculated response of an  $He^3$  detector placed in the core (upper curve) and the reflector (lower curve) of a uranium reflected assembly.

 $B^2$  in these expressions is taken as  $7.9 \cdot 10^{-3}$  cm<sup>-2</sup>, which is the mean value obtained if a in Eq. 6.1 is set equal to  $7.4 \cdot 10^4$  in groups 15 to 18. Since expression 6.1 then becomes almost constant in all low energy groups it is reasonable that an approximately exponentially decaying neutron density is obtained. The obtained buckling is equal to that of a 35.3 cm thick slab which is not an unrealistic result compared to the reflector thickness of 35 cm.

Since all volume elements in the rtE-space are coupled to each other through diffusion, fissioning and down-scattering, the slowest decaying group will eventually determine the over all decay of the neutron flux. When this decay is reached spectral equilibrium will develop all over the

system. From the group values of  $v(\Sigma_{rem}^{+DB^2})$  plotted in Fig. 6 it is seen

that the slowest rate of removal will occur in group 22. In this energy group both the leakage and the scattering out of the group are almost negligible compared to the absorption. Since this will persist even at lower energies and the capture cross section here is proportional to 1/v, the same decay constant should have been reached if more groups had been added. The expected  $\alpha$  is thus a true eigenvalue not dependent on group structure. At these energies thermal up-scattering starts to be of importance but then the absorption dominates the removal even more and the above statement is still valid. The assumption that the lowest energy group eventually determines the decay is supported by the result of a stationary  $S_{1}$ -calculation shown as straight lines in Figs. 1-3. The last group here reached from 0.2 to 8 eV giving a  $(v_{\Gamma_{rem}}^{+}+DB^2)$  of 7.6.10<sup>4</sup> s<sup>-1</sup> which should be compared to the obtained a of 7.43.10<sup>4</sup> s<sup>-1</sup>.

The determination of the equilibrium spectrum is equivalent to solving the static Boltzmann equation with all absorption cross sections decreased by a/v. This means that the effective removal from the reflector almost vanishes in groups 15-22 and that the spectrum here will be proportional to 1/E in contrast to the critical spectrum which decreases rapidly with energy. This decrease in effective absorption explains why the low energy neutrons play a much greater role in the pulsed experiment than in the steady state reactor.

The fact that the core neutrons and the high energy reflector neutrons have not reached an exponential decay even at the end of the studied time interval, although they seem to approach one, also supports the statement that the lowest energy group in the reflector eventually will determine the decay. In the high energy groups spectral equilibrium is reached much earlier. An attempt to fit two exponentials to the calculated core decay has been made without success showing the inadequacy of the two-point model in this case. This is also to be expected since the leakage probability from the reflector to the core is changing as long as the spectrum changes. This is contradictory to the assumption that all parameters entering the model are time independent. It is also evident that no unique neutron life-time can be defined and measured as long as the spectrum changes.

The time-dependent code has also been tested on a more normal reactor a core with the same composition surrounded by a natural uranium blanket. This assembly was also 3.5 % subcritical. As is seen in Fig. 7 the same exponential decay is rapidly attained throughout the reactor making it possible to use the point reactor model.

### 7. Conclusions

The discussion in the preceding sections shows that it is possible to describe quantitatively the pulsed source experiments also in systems with light reflectors with the time-dependent  $S_N$  code and with the cross section set calculated by the SPENG program. The latter part of the decay is dominated by low-energy cross sections not tested in criticality calculations, but also by the perhaps less interesting reflector properties.

It is obvious that no spectrum equilibrium is reached during the time when enough neutrons are left in the system to make measurements possible. This makes the simple one- and two-point models inadequate and parameters such as neutron life-time dependent of time and impossible to measure with the pulsed source method. A difference between critical spectrum and prompt a-mode spectrum will persist even in cases where the point reactor model is applicable giving a possible explanation to the discrepancy between calculated and experimental neutron life-times reported for instance by Brunson et al. [8].

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## DISCUSSION

B.I. SPINRAD: It seems to me that  $\alpha$  is determined by the timescale on which measurements are made. After an appropriate time one sees the decays dominated by delayed neutrons; if one waits a shorter time one sees the reflector-spectrum neutrons, which are a pseudo-thermal group of neutrons acting as a pseudo-delayed group; after an even shorter time, one sees core characteristics. Does this not mean that all these  $\alpha$  values, in themselves, contain a rather small amount of information?

J. KOCKUM: I agree that we do not get out any parameters that are characteristic of the stationary fundamental mode of the core. On the other hand, I believe the paper demonstrates that we are able to describe this particular experiment.

B.I. SPINRAD: The group of low-energy neutrons in the reflector, acting as a delayed source, introduces only a reflector flux and a small fission peak at the core edges. The shape of this function is very far from the fundamental mode of the reactor, so it is not at all surprising that one does not find it in a calculation which essentially emphasizes the fundamental mode and its decay.

J. KOCKUM: I don't think that one can describe the returning reflector neutrons as a single new group of delayed neutrons, since their energy distribution changes with time. The stationary calculations reported show that it is possible to calculate the final decay without recourse to the timedependent code. This decay, however, is as you say not a characteristic of the core; on the other hand it should be taken into account since eventually the flux at the core centre slowly follows the same decay – even if this is not measurable.

## PROBABILITY DISTRIBUTION OF NEUTRON COUNTS FOR THE p-METHOD IN REACTOR NOISE ANALYSIS A numerical calculation

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## Abstract

PROBABILITY DISTRIBUTION OF NEUTRON COUNTS FOR THE p-METHOD IN REACTOR NOISE ANALYSIS: A NUMERICAL CALCULATION. The p-method in reactor noise analysis is the technique, proposed by Zolotukhin and Mogil'ner, of measuring the probabilities  $p_k$  for detecting k neutrons in a certain time interval. Reactor parameters may be derived from the probability distributions. Because of its simplicity and high speed this method seems to be very attractive for application in fast reactors. It was used by Mogil'ner in the USSR BR-6, and a probability analyser has been built in the framework of the Karlsruhe fast breeder project.

The distribution can be used in different ways, for example by using only  $p_0$  or the variance. It is more attractive, however, to use the complete distribution, and to compare it to a theoretical calculated distribution. Up to now for such a calculation only approximate distributions were available (negative binomial distribution and an asymptotic distribution for long time intervals). The present paper describes a numerical technique for a more exact distribution. It is a calculation for the case for which a probability generating function has been derived by several authors. A point reactor model is assumed, with neglect of delayed neutrons and some very improbable branching processes.

The procedure is presented for obtaining numerically the distribution from the generating function. It is a multiple recurrence calculation that has been programmed in Algol-60 for the El-X8 computer.

Several examples of the calculated distributions are given. A comparison is made with the approximating distributions, and conclusions are obtained on the range of validity of the approximations. Moreover the calculated distributions are compared to some experimental ones, and the agreement is found to be excellent. The problem of fitting theoretical to experimental distributions is discussed.

## 1. INTRODUCTION

The p-method in reactor noise analysis is the technique proposed by Mogil'ner and Zolotukhin [1] of measuring the probabilities  $p_k$  for detecting k neutrons in a time interval T, for different values of T. Reactor parameters may be derived from these probability distributions.

The technique is very promising as a fast measuring method. By means of rather simple electronics one can measure  $p_k$  (especially  $p_o$ ) at high speed. It thus seems to be very attractive for application in fast reactors.

Experimentally the p-method has been used in the USSR reactors BR-6 (prompt neutron decay constant at critical  $\alpha_c = 5000 \text{ s}^{-1}$ ) and PF-4 ( $\alpha_c \simeq 200 \text{ s}^{-1}$ ) [2], in the French reactor ALECTO ( $\alpha_c = 50 \text{ s}^{-1}$ ) [3], in the Italian ROSPO [4], while in the framework of the Karlsruhe fast breeder project a probability analyser has been built [5] for the experimental determination of  $p_k$ .

Zolotukhin and Mogil'ner [6] recognized that in first approximation the distribution  $p_k$  is a negative binomial distribution (or Polya distribution),

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and they have shown experimentally that a negative binomial distribution is in good agreement with an experiment. Later Pacilio [4] came to the same conclusion, even for larger detector efficiencies.

Another approximate distribution has been given in [7], which is good for long time intervals. We will refer to this distribution as the asymptotic distribution. A summary of the approximations can be found in [8].

Pål [9], Zolotukhin and Mogil'ner [7] and Dalfes [10] have derived a probability generating function for a more exact probability distribution. This rather complicated function has not been used up to now for a calculation of  $p_K$ . In this paper we derive a numerical technique (a multiple recurrence process) for such a calculation. Results will be compared with the approximate distributions mentioned above.

## 2. TAYLOR EXPANSION OF THE GENERATING FUNCTION

The generating function is:

$$\pi_{\rm T}({\rm x}) = \left\{ \frac{(1+\phi)^2 e^{-{\rm a}(1-\phi)} - (1-\phi)^2 e^{-{\rm a}(1+\phi)}}{4\phi} \right\}^{-{\rm A}}$$
(1)

with:

$$a = \frac{1}{2} \alpha T$$

$$A = \frac{2m}{\alpha z T}$$

$$\phi = \sqrt{1 + 2z (1 - x)}$$
(2)

Here:

- $\alpha$  = prompt neutron decay constant
- $z = variance-to-mean ratio minus one, for T \rightarrow \infty$  (neglecting delayed neutrons)
- m = mean number of counts in the time interval T

Since the generating function is defined by

$$\pi_{\mathrm{T}}(\mathbf{x}) = \sum_{k=0}^{\infty} \mathbf{p}_{k} \mathbf{x}^{k}$$
(3)

the problem is to find the Taylor series expansion of the function (1). We make this expansion in the following 8 steps. Step 1. The expansion coefficients of

$$\phi = \sum_{k=0}^{\infty} c_k x^k$$
 (4)

are:

$$c_{k} = -\left(\frac{z}{1+2z}\right)^{k} \frac{(2k-3)!!}{k!} \sqrt{1+2z}$$
 (5)

They can be calculated by the recurrence relation

$$c_{m} = c_{m-1} B \frac{2m-3}{m}$$
 with  $B = \frac{z}{1+2z}$  (6)  
 $c_{0} = \sqrt{1+2z}$ 

Step 2. For the function  $1/\phi$  we have the expansion:

$$\frac{1}{\phi} = -\sum_{k=0}^{\infty} \frac{c_k}{1+2z} (2k-1) x^k$$
(7)

Step 3. Combining steps 1 and 2:

$$\phi + \frac{1}{\phi} = \sum_{k=0}^{\infty} g_k x^k$$
(8)

with:

$$g_{m} = 2 c_{m} \frac{1 - m + z}{1 + 2z}$$
 (9)

Step 4. A recurrence relation for the coefficients  $d_k$ , defined by

$$e^{a\phi} = exp\left(a \sum_{k=0}^{\infty} c_k x^k\right) \stackrel{\text{def}}{=} \sum_{k=0}^{\infty} d_k x^k$$
(10)

is found by differentiating both sides of the equation:

$$a \sum_{k=0}^{\infty} k c_k x^{k-1} \sum_{l=0}^{\infty} d_l x^l = \sum_{l=0}^{\infty} k d_k x^{k-1}$$
(11)

By writing m = 1 + k, and by changing the order of the summations, the left-hand side can be written:

$$a\sum_{m=0}^{\infty} x^{m-1} \left\{ \sum_{k=0}^{m} k c_k d_{m-k} \right\}$$

From this follows the recurrence relation:

$$d_{m} = \frac{a}{m} \sum_{k=1}^{m} k c_{k} d_{m-k}$$
, with  $d_{0} = e^{aC_{0}}$  (12)

Step 5. In the same way:

 $\mathbf{e}^{-\mathbf{a}\phi} = \sum_{k=0}^{\infty} \mathbf{e}_k \mathbf{x}^k$ (13)

wi**t**h

 $e_m = -\frac{a}{m} \sum_{k=1}^{m} k c_k e_{m-k}; \quad e_0 = e^{-ac_0}$  (14)

$$\left(\frac{1}{\phi} + \phi\right) \left(e^{a\phi} - e^{-a\phi}\right) = \sum_{k=0}^{\infty} h_k x^k$$
(15)

By substituting (8), (10) and (13), collecting terms with the same power  $x^k$ , one easily finds:

$$h_m = \sum_{k=0}^{m} (d_k - e_k) g_{m-k}$$
 (16)

Step 7. The expansion coefficients of:

$$\frac{(1+\phi)^2 e^{-a(1-\phi)} - (1-\phi)^2 e^{-a(1+\phi)}}{4\phi} \quad \underline{\det} \sum_{k=0}^{\infty} b_k x^k$$
(17)

are now seen to be given by

$$b_m = \frac{e^{-a}}{4} \{h_m + 2(d_m + e_m)\}$$
 (18)

$$\pi_{\mathrm{T}}(\mathbf{x}) = \sum_{k=0}^{\infty} \mathbf{p}_{k} \mathbf{x}^{k} = \left(\sum_{k=0}^{\infty} \mathbf{b}_{k} \mathbf{x}^{k}\right)^{-\mathrm{A}}$$
(19)

By differentiating both sides of the equation, one finds:

$$\sum_{l=0}^{\infty} 1p_{l}x^{l-1} \sum_{k=0}^{\infty} b_{k}x^{k} = -A \sum_{k=0}^{\infty} p_{l}x^{l} \sum_{k=0}^{\infty} kb_{k}x^{k-1}$$
(20)

Taking k + l = m, and changing the order of the summations, leads to:

$$\sum_{k=0}^{m} b_{k} p_{m-k}(m-k) = -A \sum_{k=0}^{m} k b_{k} p_{m-k}$$
(21)

which gives the recurrence relation:

$$p_{m} = -\frac{1}{m} \sum_{k=1}^{m} b_{k} p_{m-k} (m - k + Ak) / b_{0},$$
 (22)

together with

$$p_0 = b_0^{-A}$$
 (23)

The numerical process defined by the equations (6), (9), (12), (14), (16), (18) and (22) was programmed in the form of an ALGOL-60 procedure. The text of this procedure is given in the Appendix.

## 3. RESULTS OF CALCULATIONS AND EXPERIMENTS

A computer programme for the Electrologica EL-X8 computer was made, which calculates the distribution, and compares it with the following distributions:

(i) Poisson distribution;

(ii) Polya or negative binomial distribution; and

(iii) The asymptotic distribution;

all of them computed for the same set of reactor parameters.

The calculations were made for situations where experimental data were also available. The experimental distributions were measured in the LFR at Petten. The experimental technique and analysis are described in [11].

It was found that, in all cases, the experiments were in close agreement with the calculated distributions. The Poisson distribution always showed large deviations. The Polya and asymptotic distributions sometimes agreed with the "exact" one, sometimes not. This is illustrated in Figs 1 to 4.

In Fig. 1 the two approximations are good, in Fig. 2 only the Polya distribution is a good approximation, while in Fig. 3 only the asymptotic distribution is good. Neither approximation is good in Fig. 4.

## 4. DISCUSSION

From the results shown in Figs 1-4 the following conclusions are obtained. As was to be expected, the asymptotic distribution is only a good approximation in the case of rather long time intervals. For rather high values of z the Polya distribution becomes a bad approximation. This is especially so for low values of k, most notably for  $p_0$ . Figures 3 and 4 show that one has to be somewhat careful if one wants to use the Polya



FIG. 1. Calculated probability distribution for  $\alpha = 444 \text{ s}^{-1}$ , z = 0.601, T = 0.020 s,  $C = 335 \text{ s}^{-1}$ .



FIG.2. Calculated probability distribution for  $\alpha = 61.3 \text{ s}^{-1}$ , z = 49.4, T = 0.002 s,  $C = 186.8 \text{ s}^{-1}$ .

distribution, as has been strongly recommended in Refs [4, 12]. The technique of this paper gives a means for investigating the limitations.

We have not tried to fit the theoretical "exact" distributions to the experimental ones. The parameters for the calculated distributions have been obtained from other types of analysis of the same measurements. It is possible in principle to make such a fit, for example by the method of least squares. Then, for the usual iterative least-squares analysis, the derivatives  $\partial p_k / \partial \alpha$  and  $\partial p_k / \partial z$  are needed. They can be found by a recursion calculation, similar to that in section 2, for  $\partial \pi_T / \partial \alpha$  and  $\partial \pi_T / \partial z$ , which are rather complicated functions. What is to be expected



FIG.3. Calculated probability distribution for  $\alpha = 450 \text{ s}^{-1}$ , z = 6.24, T = 0.038 s,  $C = 179.2 \text{ s}^{-1}$ .



FIG.4. Calculated probability distribution for  $\alpha = 54.2 \text{ s}^{-1}$ , z = 23.7, T = 0.015 s,  $C = 532 \text{ s}^{-1}$ .

from such an analysis? In many cases the distribution is close to a Polya distribution. The parameters  $\alpha$  and z are contained in this distribution as only one parameter:

$$z\left(1 - \frac{1 - e^{-\alpha T}}{\alpha T}\right)$$
(24)

It is to be expected, therefore, that a fit of the theoretical "exact" distribution to the experimental one will yield a reliable value for the parameter (24), but not for  $\alpha$  and z separately. For such a determination distributions for at least two values of T are necessary. The least-squares analysis of two or more complete distributions at the same time will be difficult.

#### APPENDIX

The text of the ALGOL-procedure for the calculation of the probability distribution is given below. The input quantities of the procedure are:

a = prompt neutron decay constant  $\alpha$ ;

z =the parameter defined after (2.2);

C = count-rate;

T = length of time interval.

The distribution  $p_k$  is delivered in the array p[1:max].

procedure EXACT(a,z,C,T,p,max); value a; z; C, T; real a, z, C, T; integer max; array p; begin real x, A, B, D, h, y; integer m,k,m1; array c,g,d,e,b[0:max]; real procedure SUM(1,h,k,t1); value k; integer i; h, k; real ti; begin real s; **s:= 0; 1:= h; goto test;** next: s:= s+t1; 1:= 1+1; test: if i k then goto next; SUM:= s end SUM;  $x := a \times T/2; A := 2 \times C/(a \times z);$ y := exp(-x);B:= z/(1+2xz);c[0]:=\_sqrt(1+2xz);  $g[0] := 2 \times (1+z)/c[0];$  $d[0]:= \exp(x \times c[0]); e[0]:= 1/d[0];$  $D:= b[0]:= y \times (g[0] \times (a[0]-e[0]) + 2 \times (a[0]+e[0]))/4;$ h:= p[0]:= b[0]/(-A);m:= m1:= 0; C:= C×T; for m := m+1 while  $(m < C \lor h > m-6) \land m < max$ do begin  $c[m] := c[m-1] \times B \times (2 \times m-3)/m;$  $g[m] := 2 \times c[m] \times (1-m+z)/(1+2 \times z);$  $d[m] := SUM(k, 1, m, k \times c[k] \times d[m-k]) \times x/m;$  $e[m]:=-SUM(k,1,m,k\times [k]\times [m-k])\times (m;$ h:= SUM(k,0,m,(d[k]-e[k])\times g[m-k]);  $b[m] := y \times (h+2 \times (d[m]+e[m]))/4;$  $h := p[m] := -SUM(k, 1, m, (m-k+A \times k) \times b[k] \times p[m-k])/(D \times m);$ m1:= m1+1 ; end; for m:= m1 step 1 until max do p[m]:= 0; max:= m1

end EXACT;

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## DISCUSSION

D. STEGEMANN: I should like to point out that by his method Mr. Turkcan was calculating probability distributions which we measured here in STARK and SNEAK by means of a probability analyser, and that agreement between theory and experiment was found to be very good. We gratefully acknowledge the work done on these calculations and feel that real progress has been made in the application of this method by virtue of Mr. Turkcan's theoretical treatment. What is still lacking, however, is a thorough treatment of statistical errors for the calculated probabilities, and we hope that Mr. Turkcan will go on with this encouraging treatment to solve this error problem.

# MODAL EFFECTS IN THE TRIPLE-CORRELATION REACTOR-NOISE EXPERIMENT

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## Abstract

MODAL EFFECTS IN THE TRIPLE-CORRELATION REACTOR-NOISE EXPERIMENT. Recently the point reactor theory for detection of three correlated counts has been given by the author elsewhere. Two types of family-trees for the three related neutrons have to be distinguished: in one the tree twice branches in two; in the other type there is a threefold splitting. This last effect is most interesting physically, since it depends on a new reactor parameter, which permits the determination of delayed neutron fraction, generation time (separately) and fission rate, without the need for any static experiment. The effect, however, is very weak. It has been shown that the special triple correlation function  $n(t)\{n(t)-n(t+\tau)\}n(t+\tau)$ (where n(t) = count-rate of a neutron detector) only depends on the threefold splitting effect, at least for point reactor theory. The determination of this quantity is a typical fast reactor experiment, since only then: (a) there are no interfering delayed neutron effects; (b) the measuring time can be kept within reasonable limits. In the present paper the theory is extended to include space dependence. The triple correlation function is generalized to:  $n(t)\{n(t+e)-n(t+e+\tau)\}n(t+2e+\tau)$ , permitting better discrimination against higher modes.

The theory uses the Green's function approach that has been used by several authors for the calculation of second order moments. Two main conclusions are obtained: (a) A correction factor is found for the new reactor parameter, mentioned above, containing integrals over multiple products of fission density and neutron importance; (b) The higher modes of the twofold double-splitting tree are found to produce in the triple correlation function a contribution which is no longer zero but may eventually exceed the desired effect. It will be calculated for a few simple cases.

An experiment was performed for measuring the triple correlation functions, by coupling a reactor online to a digital computer. Since a thermal reactor was used, only very rough results could be obtained, in spite of the long measuring time. The spatial effect was clearly observed, not the threefold splitting effect.

## 1. INTRODUCTION

In a recent paper [1] a theory was given on third-order moments and threefold correlations in the neutron noise of a stationary reactor for the point reactor model.

Three neutrons, detected by a neutron counter (or three counters), may be correlated if the fission trees to which they belong, somewhere in the past originate from a common fission. If one traces back these fission trees one may find, of course,

(a) No correlation at all;

(b) Two correlated and one uncorrelated neutron chain;

(c) Three correlated chains.

In this paper we will only consider the threefold correlated events. They may belong to two different types of fission tree, shown in Fig.1.

The fundamental quantity in the theory is the probability for detecting a neutron in dt<sub>1</sub> at t<sub>1</sub>, another neutron in dt<sub>2</sub> at t<sub>2</sub> and a third one in dt<sub>3</sub> at t<sub>3</sub>. We will denote this probability for process a of Fig.1 by  $P_a(t_2-t_1, t_3-t_1)$ , and for process b by  $P_b(t_2-t_1, t_3-t_1)$ .  $P_b$  can be split into three



FIG.1. Fission trees for threefold correlation. The solid lines (-) represent fission chains, crosses  $(\times)$  are fission events, and circles (O) detected neutrons.

different contributions  $P_{b1}$ ,  $P_{b2}$  and  $P_{b3}$ , depending on whether the count labelled A (see Fig.1) is the first, second or third count.

It has been found in [1], for the point reactor model, with neglect of delayed neutrons:

$$P_{a}(u, v) = \frac{1}{3} C \alpha^{2} Z_{2} e^{-\alpha(u+v)}$$
(1)

$$P_{b}(u, v) = \frac{1}{2} C \alpha^{2} Z_{1}^{2} e^{-\alpha v}$$
(2)

with:

C = mean count-rate of the detector

 $\alpha$  = prompt neutron decay constant and with:

$$Z_1 = \epsilon \frac{\overline{\nu(\nu-1)}}{\overline{\nu^2}} \cdot \frac{1}{\alpha^2 \Lambda^2}$$
(3)

$$Z_2 = \epsilon^2 \frac{\overline{\nu(\nu-1)(\nu-2)}}{\overline{\nu}^3} \cdot \frac{1}{\alpha^3 \Lambda^3}$$
(4)

where:

 $\epsilon$  = detector efficiency = ratio of count-rate C and fission rate F;

 $\nu$  = number of neutrons per fission;

 $\Lambda$  = generation time.

 $Z_1$  is the same parameter that can be determined in the usual twofold correlation experiments (Rossi- $\alpha$ , Feynman- $\alpha$ , etc., see [2]).

The possibility of measuring  $Z_2$  is interesting. It permits the determination of  $\epsilon$  (and from that the fission rate F or the reactor power) and of  $\alpha \Lambda$  (=  $\beta - \rho$  = prompt reactivity) separately, provided the statistics of  $\nu$  are known with sufficient accuracy. For a reactor close to delayed criticality the delayed neutron fraction  $\beta$  can be obtained directly.

Now, ingeneral, it is much more likely that three detected neutrons belong to a fission tree of type b than to type a. Experimentally the tree-a effect will nearly always be obscured by the tree-b effect. In [1], however, a promising third-order moment was found:

$$n(t)\{n(t) - n(t + \tau)\}n(t + \tau)$$
(5)

where n(t) represents the detector count-rate. For this particular moment can be written:

## $\overline{n(t)\{n(t) - n(t + \tau)\}} n(t + \tau) = \{P_a(0, \tau) - P_a(\tau, \tau)\} + \{P_b(0, \tau) - P_b(\tau, \tau)\}$ (6)

By substituting (1) and (2), the term with  $P_b$  is seen to cancel, and only the tree-a effect is left. Measurement of (5), therefore, might yield  $Z_2$ . In [1] the effect of delayed neutrons has also been calculated. The delayed neutron contribution to  $P_b$  can disturb the prompt neutron effect in  $P_a$ , but this effect is negligible in fast systems. The determination of (5) was concluded to be a typical fast reactor experiment for two reasons:

(a) In this case the delayed neutrons are not interfering;

(b) Since the time scale of the experiment is determined by  $\alpha$ , and a very large number of intervals  $1/\alpha$  will be needed, a reasonable measuring time is only to be expected in fast systems.

The effects of space and energy dependence have not been taken into account in [1]. This will be the subject of the present paper. The need for the evaluation of this effect will appear in the next section.

## 2. EXPERIMENT

Although the experiment is typically suited to fast reactors, it was nevertheless tried in a thermal system, viz. the Argonaut reactor LFR at Petten, in December 1966. A very long measuring time and poor accuracy were to be expected. Here we will give a brief description and some results of the experiment, since on the one hand the experiment shows one possible way of obtaining this information, and on the other hand the results have given rise to the evaluation of modal effects in this paper.

The experiment was as follows. The counts from a  $BF_3$ -counter in the reactor were fed to a binary scaler. The scaler was read and re-set every 2 ms (time interval T), and the contents (numbers  $n_i$ ) were transferred directly to the Electrologica EL-X8 computer. Here the quantity

$$\frac{1}{N} \sum_{i=1}^{N} n_i (n_i - n_{i+j}) n_{i+j}$$
(7)

was calculated, together with the twofold correlation function

$$\frac{1}{N} \sum_{i=1}^{N} n_{i} n_{i+j} - \overline{n^{2}}$$
(8)

It was possible to calculate the functions (7) and (8) in real time for at most 24 different values of j at the same time. Once in about 7 min the measurement was interrupted in order to print intermediate results. Measurements were taken for about 75 min.

Detector position	C (counts/s)	α (s <sup>-1</sup> )	Z <sub>1</sub>	A/Z <sup>2</sup>
VN	1100	53.7	42.7	$(2.79 \pm 0.17) \times 10^{-2}$
VC	650	54.4	14.2	$(2.49 \pm 0.30) \times 10^{-2}$





FIG.2. Third-order moment measured in VN.



FIG. 3. Third-order moment measured in VC.

The results have been analysed by fitting the auto-correlation function (8) to:

$$\frac{1}{2}CT^2 \alpha Z_1 e^{-\alpha jT}$$
(9)

thus obtaining  $\alpha$  and  $Z_1$ . Then (7) was fitted to

$$CT^{3}\alpha^{2}Ae^{-\alpha\tau}(1-e^{-\alpha\tau})$$
(10)

When the desired effect is observed, A is equal to  $\frac{1}{3}Z_2$ , and  $A/Z_1^2$  is equal to  $(\beta - \rho)/4$ .

Results are presented in Table I and Figs 2 and 3. Measurements were made at two different detector positions at the same reactivity (about -8 cents). If threefold correlations are detected (either from tree-a or tree-b), the quantity  $A/Z_1^2$  should be independent of the position (i.e. A proportional to  $\epsilon^2$ ). This was in fact observed. However, we know that, for this reactor,  $\beta \approx 0.007$ . The expected  $A/Z_1^2$  then is  $0.19 \times 10^{-2}$ . The experimental quantity is larger by a factor of about 14. The most likely explanation is that this is due to the only effect that has been neglected so far: higher modes of tree-b, which may introduce a contribution that is not distinguishable from the tree-a effect.

Part of the higher mode effect may be due to the fact that two of the three points in the triple correlation function coincide. This contribution is eliminated by spacing the three points. It can easily be shown that the following generalization of (5):

$$\overline{n(t)\{n(t+\sigma) - n(t+\sigma+\tau)\}n(t+2\sigma+\tau)}$$
(11)

possesses the property of only depending on threefold correlated counts, and in the point reactor model on tree-a only. Also, this quantity was determined in the on-line experiment. The result was roughly the same as for the measurement with  $\sigma = 0$ , but much more inaccurate. The generalized moment (11) will be used in the subsequent sections.

## 3. THE MODAL THEORY

## 3.1. Definitions, etc.

Green's function approach will be used, as it was used for the first time for space dependent problems by Matthes [3]. We will closely follow the work of Otsuka and Iijima [4].

Space and energy dependence will be taken into account. These coordinates together will be denoted as one phase space co-ordinate x. The reactor is stationary in time. Only prompt neutron multiplication will be considered.

The following Green's function is needed:

 $\pi(\mathbf{x}_1, \mathbf{x}_0, \mathbf{t}_1-\mathbf{t}_0)$  = neutron density at  $\mathbf{x}_1$ , and at time  $\mathbf{t}_1$ , following one neutron born at  $\mathbf{x}_0$  and at time  $\mathbf{t}_0$ . Our functions will be expanded in eigenfunctions. We use  $\omega_p$ -modes [5] (prompt neutron production only). The neutron density eigenfunction will be denoted by  $\phi_k$ , the adjoint eigenfunctions by  $\phi_k^*$ , the eigenvalues by  $-\alpha_k$  (the fundamental  $\alpha_0$  is our previously defined prompt neutron decay constant  $\alpha$ ). Then:

$$\pi(\mathbf{x}_1, \mathbf{x}_0, \mathbf{t}_1 - \mathbf{t}_0) = \sum_{k} \phi_k(\mathbf{x}_1) \phi_k^*(\mathbf{x}_0) e^{-\alpha_k(\mathbf{t}_1 - \mathbf{t}_0)}$$
(12)

where the eigenfunctions are so normalized that:

$$\int \phi_{\nu}^{*}(\mathbf{x}) \phi_{\nu}(\mathbf{x}) d\mathbf{x} = 1 \tag{13}$$

The fission process will be described by the operator  $\mathscr{T}$  (in general the product of fission cross-section and normalized fission spectrum), which means that a neutron density N(x) produces a fission density in phase space:

$$\mathbf{F}(\mathbf{x}) = \mathscr{F} \mathbf{N}(\mathbf{x}) \tag{14}$$

Henceforth N(x) and F(x) will be used for the stationary distributions.

We will have three neutron detectors in the reactor. We start by assuming monoenergetic point detectors at  $x_i$  in phase space. They have detection probabilities  $\eta_i$  (i = 1, 2, 3), which means that a neutron density N(x) produces a count-rate

$$C_i = \eta_i N(x_i) \tag{15}$$

For more realistic detectors we will have to integrate our later results over some part of the phase space. As far as point detector results will be found to depend only on the stationary  $C_i$ , we are permitted to apply them for finite detectors as well.

## 3.2. Calculation of $P_a(u, v)$

 $P_a$  is found by means of the following list of probabilities:

- (i) Probability for fission in  $dx_0$  at  $x_0$  and in  $dt_0$  at time  $t_0$ :  $F(x_0)dt_0dx_0$
- (ii) Probability for production of  $\nu$  prompt neutrons:  $p_{\nu}$
- (iii) Probability for detection of a descendant at  $x_1$ ,  $t_1$ :  $\eta_1 \nu \pi$  ( $x_1$ ,  $x_0$ ,  $t_1 t_0$ )dt<sub>1</sub>
- (iv) Probability for detecting another descendant at  $x_2$ ,  $t_2$ :  $\eta_2(\nu-1)\pi(x_2, x_0, t_2-t_0)dt_2$
- (v) Probability for detecting a third descendant at  $x_3$ ,  $t_3$ :  $\eta_3(\nu 2)\pi(x_3, x_0, t_3 t_0)dt_3$

These probabilities are multiplied, Eq. (12) is used, and the integrations over space and time are carried out. We are only interested in a firstorder approximation. In the phase space integrals the contributions with  $\phi_0$  and  $\phi_0^*$  appear to be dominating: we only take them into account. Further we assume that the stationary density N(x) is approximately proportional to  $\phi_0(x)$ . Then:

$$F(x) = K \mathscr{F} \phi_0(x)$$

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(16)

. 31\*

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$$\mathbf{C}_{\mathbf{i}} = \mathbf{K} \eta_{\mathbf{i}} \phi_0(\mathbf{x}_{\mathbf{i}}) \tag{17}$$

In this fundamental mode approximation, the probability  $P_a$  is immediately seen to be equal to:

$$P_{a}(\mathbf{u}, \mathbf{v}) = \eta_{1} \eta_{2} \eta_{3} \phi_{0}(\mathbf{x}_{1}) \phi_{0}(\mathbf{x}_{2}) \phi_{0}(\mathbf{x}_{3}) \overline{\nu(\nu - 1)(\nu - 2)} \int \phi_{0}^{*}(\mathbf{x}_{0})^{3} F(\mathbf{x}_{0}) d\mathbf{x}_{0}$$

$$\times \int_{-\infty}^{t_{1}} e^{-\alpha(t_{1} - t_{0}) - \alpha(t_{2} - t_{0}) - \alpha(t_{3} - t_{0})} dt_{0}$$

$$= \frac{C_{1} C_{2} C_{3}}{3\alpha K^{2}} \int \phi_{0}^{*3} \mathscr{F} \phi_{0} d\mathbf{x} \cdot e^{-\alpha(u + v)}$$
(18)

Using the definitions:

Total fission rate 
$$F = \int F(x) dx = K \int \mathscr{F} \phi_0 dx$$
 (19)

Generation time 
$$\Lambda = \frac{1}{\overline{\nu} \int \phi_0^* \mathscr{F} \phi_0 \, dx}$$
 (20)

( $\overline{\nu}$  = Average number of prompt plus delayed neutrons per fission);

Efficiency 
$$\epsilon_i = C_i / F$$
 (21)

one obtains:

$$P_{a}(u, v) = \frac{1}{3}C_{1}\alpha^{2}Z_{2}f_{2}e^{-\alpha(u+v)}$$
(22)

Here  $Z_2$  is the quantity defined in (4), but now with  $\epsilon_2 \epsilon_3$  instead of  $\epsilon^2$ .

The effect of space and energy dependence is expressed in the factor  $f_2$  (a correction factor with respect to the point reactor model result), given by:

$$f_2 = \frac{\int \phi_0^{*3} \mathscr{F} \phi_0 \, dx \cdot \{\int \mathscr{F} \phi_0 \, dx\}^2}{\{\int \phi_2^* \mathscr{F} \phi_0 \, dx\}^3}$$
(23)

## 3.3. Calculation of $P_b(u, v)$

The list of probabilities for  $P_{b1}$  is as follows:

- (i) Probability for fission at  $t_0$ ,  $x_0$ :  $F(x_0) dt_0 dx_0$
- (ii) Probability for production of  $\nu$  neutrons:  $p_{\nu}$
- (iii) Probability of detecting a descendant at  $t_1$  with counter 1:  $\eta_1 \nu \pi(x_1, x_0, t_1 t_0)$
- (iv) Probability that another descendant gives fission at  $t_4$ ,  $x_4$ :  $(\nu - 1) \mathscr{F}_4 \pi(x_4, x_0, t_4 - t_0); (\mathscr{F}_4 \text{ means hat } \mathscr{F} \text{ operates on } x_4)$
- (v) Probability for  $\mu$  neutrons produced at  $t_4$ :  $p_{\mu}$
- (vi) Probability for detecting a descendant at  $t_2$  with counter 2:  $\eta_2 \mu \pi(x_2, x_4, t_2 - t_4)$
- (vii) Probability for detecting another descendant at  $t_3$  with counter 3:  $\eta_3(\mu-1)\pi(x_3, x_4, t_3-t_4)$

Again we use the expansion (12), but this time not the fundamental mode approximation. This results in:

$$P_{b1}(u, v) = \sum_{\substack{k \ell m n}} S^{1}_{k \ell m n} T^{1}_{k \ell m n}$$
(24)

where the time factor - exactly as in [1] - is given by:

$$\Gamma_{k\ell mn}^{1} = \int_{0}^{\infty} ds \int_{0}^{s+u} dt e^{-\alpha_{k}s - \alpha_{\ell}t - \alpha_{m}(u+s-t) - \alpha_{n}(v+s-t)}$$
$$= \frac{e^{-(\alpha_{\ell} - \alpha_{n})u - \alpha_{n}v}}{(\alpha_{m} + \alpha_{n} - \alpha_{\ell})(\alpha_{k} + \alpha_{\ell})} - \frac{e^{-\alpha_{m}u - \alpha_{n}v}}{(\alpha_{m} + \alpha_{n} - \alpha_{\ell})(\alpha_{k} + \alpha_{\ell} + \alpha_{m})}$$
(25)

The phase space factor  $S^{1}_{k\ell mn}$  is found to be:

$$\mathbf{S}_{k\ell mn}^{1} = \eta_{1} \phi_{k}(\mathbf{x}_{1}) \cdot \eta_{2} \phi_{m}(\mathbf{x}_{2}) \cdot \eta_{3} \phi_{n}(\mathbf{x}_{3}) \cdot \overline{\nu(\nu-1)}^{2} \cdot Q_{k\ell} \mathbf{R}_{mn\ell}$$
(26)

where the abbreviations:

$$\begin{cases} \phi_{k}^{*}(\mathbf{x}) \phi_{\ell}^{*}(\mathbf{x}) \mathbf{F}(\mathbf{x}) d\mathbf{x} = \mathbf{Q}_{k\ell} \\ \int \phi_{k}^{*}(\mathbf{x}) \phi_{\ell}^{*}(\mathbf{x}) \mathscr{F} \phi_{m}(\mathbf{x}) d\mathbf{x} = \mathbf{R}_{k\ell m} \end{cases}$$

$$(27)$$

have been used.

The next problem is to find  $P_{b2}(u, v)$ . An analogous evaluation as for  $P_{b1}$  - and corresponding to the calculation in [1] - yields:

$$P_{b2}(u, v) = \sum_{k \ell m n} S_{k \ell m n}^2 T_{k \ell m n}^2$$
 (28)

$$T_{k\ell mn}^{2} = \frac{e^{-\alpha_{m}u - \alpha_{n}v}}{(\alpha_{k} + \alpha_{m} + \alpha_{n})(\alpha_{m} + \alpha_{\ell})}$$
(29)

$$\mathbf{S}_{k\ell m n}^{2} = \eta_{1}\phi_{k}(\mathbf{x}_{1}) \cdot \eta_{2}\phi_{m}(\mathbf{x}_{2}) \cdot \eta_{3}\phi_{n}(\mathbf{x}_{3}) \cdot \overline{\nu(\nu-1)^{2}} \cdot \mathbf{Q}_{m\ell}\mathbf{R}_{kn\ell}$$
(30)

Similarly:

$$P_{b3}(u, v) = \sum_{k \ell m n} S^{3}_{k \ell m n} T^{3}_{k \ell m n}$$
(31)

with

$$T^{3}_{k\ell mn} = \frac{e^{-\alpha_{m}u - \alpha_{n}v}}{(\alpha_{k} + \alpha_{m} + \alpha_{n})(\alpha_{n} + \alpha_{\ell})}$$
(32)

$$S_{k\ell mn}^{3} = \eta_{1}\phi_{k}(x_{1}) \cdot \eta_{2}\phi_{m}(x_{2}) \cdot \eta_{3}\phi_{n}(x_{3}) \cdot \overline{\nu(\nu-1)}^{2} \cdot Q_{m\ell}R_{km\ell}$$
(33)

## 3.4. Approximations in $P_b(u, v)$

Now we assume that we are interested only in time intervals of the order of magnitude of  $1/\alpha$ . If then the higher modes are much more subcritical than the fundamental one, we have:

$$e^{-\alpha_{\ell} u} \simeq e^{-\alpha_{\ell} v} \simeq 0 \quad \text{for } \ell \ge 1 \tag{34}$$

Then  $T_{k\ell mn}^2$ ,  $T_{k\ell mn}^3$  and the second term of  $T_{k\ell mn}^1$  are zero, unless m = n = 0. The first term of  $T_{k\ell mn}^1$  is zero, unless  $\ell = n = 0$ . The non-zero terms left in  $P_{b1} + P_{b2} + P_{b3}$  are:

$$P_{b}(u, v) \simeq \sum_{km} \eta_{1} \phi_{k}(x_{1}) \cdot \eta_{2} \phi_{m}(x_{2}) \cdot \eta_{3} \phi_{0}(x_{3}) \cdot \overline{\nu(\nu-1)}^{2} e^{-\alpha v} \cdot \frac{Q_{k0} R_{m00}}{\alpha_{m}(\alpha_{k} + \alpha_{0})}$$

$$+ \sum_{k\ell} \eta_{1} \phi_{k}(x_{1}) \cdot \eta_{2} \phi_{0}(x_{2}) \cdot \eta_{3} \phi_{0}(x_{3}) \cdot \overline{\nu(\nu-1)}^{2} \frac{e^{-\alpha(u+v)}}{(2\alpha + \alpha_{k})}$$

$$\left\{ \frac{Q_{k\ell} R_{00\ell}}{2\alpha - \alpha_{\ell}} + \frac{2Q_{0\ell} R_{k0\ell}}{\alpha + \alpha_{\ell}} \right\}$$

$$(35)$$

The first term, proportional to  $e^{-\alpha v}$ , is the one that cancels in the moment (11). The dominating term in the sum is the term with k = m = 0. Together with the approximations (16) and (17) and the definitions (19) - (21) this term becomes:

$$\frac{1}{2}C_1\alpha^2 Z_1(\epsilon_2) Z_1(\epsilon_3) f_1^2 e^{-\alpha v}$$
(36)

where now  $f_1$  is the space and energy correction factor

$$f_{1} = \frac{\int \phi_{0}^{*} \,^{2} \mathscr{F} \phi_{0} \, dx \int \mathscr{F} \phi_{0} \, dx}{\{\int \phi_{0}^{*} \mathscr{F} \phi_{0} \, dx\}^{2}}$$
(37)

The same factor has been given in [4] for twofold correlations.

In the second term of (35) proportional to  $\{-a(u+v)\}$ , is the contribution with  $k = \ell = 0$  exactly zero. Only higher mode contributions are left. The dominating terms are for k=0,  $\ell \neq 0$  or  $k \neq 0$ ,  $\ell = 0$ . The terms  $k \neq 0$ ,  $\ell = 0$  give a factor:

$$\frac{-Q_{k0}R_{000} + Q_{00}R_{k00}}{\alpha}$$
(38)

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which will be very small. In fact, in the approximation that the stationary distribution is proportional to the fundamental mode, we have

$$Q_{k\ell} \equiv R_{k\ell 0} \tag{39}$$

(40)

(as follows immediately from the definitions (27). Then (38) is exactly zero.

The terms k=0,  $l \neq 0$  in the second term of (35) yield:

$$\sum_{\ell} \eta_1 \phi_0(\mathbf{x}_1) \cdot \eta_2 \phi_0(\mathbf{x}_2) \cdot \eta_3 \phi_0(\mathbf{x}_3) \overline{\nu(\nu-1)}^2 \cdot \frac{\mathrm{e}^{-\alpha(\mathbf{u}+\mathbf{v})}}{\alpha} \cdot \frac{(\alpha_{\ell} - \alpha)}{(\alpha_{\ell} - 2\alpha)(\alpha + \alpha_{\ell})} \cdot Q_{0\ell} \mathbf{R}_{00\ell}$$
$$= \frac{1}{2} C_1 \alpha^2 Z_1(\epsilon_2) Z_1(\epsilon_3) f_1^2 \mathrm{e}^{-\alpha(\mathbf{u}+\mathbf{v})} \cdot \mathbf{a}$$
(40)

with

$$a = \sum_{\ell \ge 1} \frac{2\alpha(\alpha_{\ell} - \alpha)}{(\alpha_{\ell} - 2\alpha)(\alpha_{\ell} + \alpha)} \cdot \frac{R_{\ell 0 0} R_{0 0 \ell}}{R_{0 0 0}^2}$$
(41)

where (39) has been used.

The final result for  $P_b$  is:

$$P_{b}(u, v) = \frac{1}{2} C_{1} \alpha^{2} Z_{1}(\epsilon_{2}) \cdot Z_{1}(\epsilon_{3}) f_{1}^{2} \{ e^{-\alpha v} + a e^{-\alpha (u+v)} \}$$
(42)

This result is to be used for the evaluation of the moment (11). Together with (22):

$$n_1(t)\{n_2(t+\sigma) - n_2(t+\sigma+\tau)\}n_3(t+2\sigma+\tau) = P(\sigma, 2\sigma+\tau) - P(\sigma+\tau, 2\sigma+\tau)$$

$$= C_{1} \alpha^{2} \{ \frac{1}{3} Z_{2} f_{2} + \frac{1}{2} Z_{1}(\epsilon_{2}) Z_{1}(\epsilon_{3}) f_{1}^{2} a \} e^{-\alpha \tau} (1 - e^{-\alpha \tau})$$
(43)

## 3.5. Discussion of the results

It is no longer true that values of  $Z_1$  and  $Z_2$  immediately yield  $\epsilon$  and  $\beta$ - $\rho$ : the factors f<sub>1</sub> and f<sub>2</sub> must be taken into account. In particular, the relation between  $Z_1$ ,  $Z_2$  and  $\beta - \rho$  [1] now must be expressed in measurable quantities as follows:

$$1.33 Z_2 f_2 / Z_1^2 f_1^2 = (\beta - \rho)g \tag{44}$$

with geometry and spectrum factor:

$$g = \frac{f_2}{f_1^2} = \frac{\int \phi_0^{*3} \mathscr{F} \phi_0 \, dx \int \phi_0^* \mathscr{F} \phi_0 \, dx}{\{\int \phi_0^{*2} \mathscr{F} \phi_0 \, dx\}^2}$$
(45)

This factor is expected to be not too far from 1.

A second conclusion is that the higher modes of case b in Fig.1 introduce a disturbing effect in the moment (11). It appears from Eq. (43) that it is not possible to remove this disturbance by suitably positioning the

detectors. In the following sections, therefore, only one detector will be considered.

It follows from the derivation that the effect is caused by the higher modes induced by the transition from the first to the second fission in tree-b of Fig.1. It depends completely on the reactor configuration. Its magnitude is expressed in the factor a. From (43) follows that the disturbance is small if

$$\frac{\frac{1}{2}Z_{1}^{2}f_{1}^{2}a}{\frac{1}{2}Z_{2}f_{2}} \simeq \frac{2a}{(\beta-\rho)g} \ll 1$$
(46)

In the next section the quantity a will be estimated.

## 3.6. Calculation of the disturbance a

3.6.1. General consideration

An order-of-magnitude estimate for the quantity a can be obtained by only considering the term l=1. If, moreover,  $\alpha_1 \gg \alpha$ :

$$\mathbf{a} \simeq \frac{2\alpha}{\alpha_1} \cdot \frac{\int \phi_1^* \phi_0^* \, \mathcal{F} \, \phi_0 \, \mathrm{dx} \int \phi_0^{*\, 2} \, \mathcal{F} \, \phi_1 \, \mathrm{dx}}{\{\int \phi_0^{*\, 2} \, \mathcal{F} \, \phi_0 \, \mathrm{dx}\}^2} \tag{47}$$

For the evaluation of this expression one needs a computer code that calculates the higher  $\omega_p$ -mode. This is a difficult problem for a fast reactor. It is possible to find higher modes by a multigroup diffusion code with the possibility for a poison search. But this process in practice only works for a low number of energy groups. For fast systems, how-ever, one is not permitted to reduce the number of energy groups to a low number; this is especially so for  $\alpha$ -calculations [6].

To gain some understanding, nevertheless, the calculation has been made for two very simple analytic reactor models: the bare homogeneous monoenergetic slab reactor, and the reflected reactor described by the two-point Cohn model. At the same time the factor g (45) will be calculated.

## 3.6.2. Bare homogeneous one-group slab

Now x is the one-dimensional space coordinate. Apart from normalization factors

$$\phi_0(\mathbf{x}) = \phi_0^*(\mathbf{x}) = \cos\frac{\pi \mathbf{x}}{2\mathbf{b}}$$
(48)

for a slab extending from x = -b to x = +b. The fission operator is a constant over the slab. The factor g now becomes:

$$g = \frac{\frac{+\pi/2}{\int} \cos^4 y \, dy \int^{+\pi/2} \cos^2 y \, dy}{\left\{\frac{+\pi/2}{\int} \cos^3 y \, dy\right\}^2} = \frac{27\pi^2}{256} = 1.041$$
(49)

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For the calculation of a (47) only <u>even</u> higher eigenfunctions must be considered (odd functions give no contribution). Then

$$\phi_1(\mathbf{x}) = \phi_1^*(\mathbf{x}) = \cos\frac{3\pi\mathbf{x}}{2\mathbf{b}}$$
 (50)

Taking also the normalization condition (13) into account, this gives (with integrations from  $-\pi/2$  to  $+\pi/2$ ):

$$a = \frac{2\alpha_0}{\alpha_1} \left\{ \frac{\int \cos^3 y \cdot \cos^2 y \cdot dy}{\int \cos^3 y \, dy} \right\}^2 \frac{\int \cos^2 y \, dy}{\int \cos^2 3y \, dy} = \frac{2}{25} \cdot \frac{\alpha_0}{\alpha_1}$$
(51)

 $\alpha_0$  and  $\alpha_1$  are estimated from

$$\alpha_{i} = \frac{1 + B_{i}^{2} L^{2} - (1 - \beta) k_{\infty}}{k_{\infty} \Lambda}$$
(52)

with diffusion length L, infinite multiplication factor  $\,k_{\,\varpi}\,$  , and geometric bucklings:

$$B_0^2 = \left(\frac{\pi}{2b}\right)^2 ; B_1^2 = \left(\frac{3\pi}{2b}\right)^2 = 9 B_0^2$$
 (53)

Then for a critical reactor, since  $k_{\infty} = 1 + B_0^2 L^2$ :

$$\alpha_0 = \beta / \Lambda$$

$$\alpha_1 \simeq \frac{8(k_\infty - 1)}{k_\infty \Lambda}$$
(54)

Then

$$a \simeq \frac{k_{\infty}\beta}{100 \left(k_{\infty} - 1\right)} \tag{55}$$

and the condition (46) becomes

$$0.02 \frac{k_{\infty}}{k_{\infty} - 1} \ll 1$$

$$k_{\infty} - 1 \gg 0.02$$
(56)

or

The more reactive the core, the smaller is the disturbing effect.

## 3.6.3. The Cohn model

This is a two-point reactor model: core and reflector both are represented by one point. These points will be labelled by c and r respectively. Then the kinetic equations for core and reflector densities, neglecting delayed neutron multiplication, are:

$$\frac{\mathrm{d}}{\mathrm{dt}} \begin{bmatrix} \mathbf{N}_{\mathrm{c}} \\ \mathbf{N}_{\mathrm{r}} \end{bmatrix} = \begin{bmatrix} (\rho_{\mathrm{c}} - \beta)/\Lambda_{\mathrm{c}} & +\kappa/\ell \\ \mu/\Lambda_{\mathrm{c}} & -1/\ell \end{bmatrix} \begin{bmatrix} \mathbf{N}_{\mathrm{c}} \\ \mathbf{N}_{\mathrm{r}} \end{bmatrix}$$
(57)

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where  $\kappa$  and  $\mu$  are coupling parameters between core and reflector, and  $\ell$  is the lifetime of neutrons in the reflector. This formulation differs from Cohn's original one [7] in that we prefer to use reactivity and generation time, rather than  $\delta k$  and lifetime. The reactivity of the whole reactor is expressed in the partial core reactivity  $\rho_c$ :

$$\rho = \rho_c + \kappa \mu \tag{58}$$

The total generation time is

$$\Lambda = \Lambda_{\rm c} + \kappa \mu \, \ell \tag{59}$$

The integrals in (45) and (47) now become summations over two points. Since  $\mathscr{F}$ , the fission operator, is zero in the reflector, the integrals containing  $\mathscr{F}$  have one non-zero term. It follows immediately that

and that

$$a = \frac{2\alpha_0}{\alpha_1} \frac{(\phi_1^*)_c(\phi_1)_c}{(\phi_0^*)_c(\phi_0)_c} \frac{\{(\phi_0^*)_c(\phi_0)_c + (\phi_0^*)_r(\phi_0)_r\}}{\{(\phi_1^*)_c(\phi_1)_c + (\phi_1^*)_r(\phi_1)_r\}}$$
(61)

where the normalization (13) has been incorporated in the formula. Now  $(-\alpha_0)$  and  $(-\alpha_1)$  are the eigenvalues of the matrix in (57):

$$\begin{vmatrix} (\rho_{c} - \beta)/\Lambda_{c} + \alpha_{i} & \kappa/\ell \\ \mu/\Lambda_{c} & -1/\ell + \alpha_{i} \end{vmatrix} = 0$$
 (62)

 $\phi_0$  and  $\phi_1$  are the eigenvectors,  $\phi_0^*$  and  $\phi_1^*$  are the eigenvectors of the adjoint matrix. They can be written

$$\phi_{i} = \begin{vmatrix} 1/\ell - \alpha_{i} \\ \mu/\Lambda_{c} \end{vmatrix} \qquad \phi_{i}^{*} = \begin{vmatrix} 1/\ell - \alpha_{i} \\ \kappa/\ell \end{vmatrix}$$
(63)

Then Eq. (61) becomes, using (59),

$$a = \frac{2\alpha_0}{\alpha_1} \frac{(\alpha_1 \ell - 1)^2}{(1 - \alpha_0 \ell)^2} \cdot \frac{\{\Lambda_c (1 - \alpha_0 \ell)^2 + \Lambda - \Lambda_c\}}{\{\Lambda_c (\alpha_1 \ell - 1)^2 + \Lambda - \Lambda_c\}}$$
(64)

We try to approximate this expression for

(i) Weak coupling, short reflector lifetime; and(ii) A system close to critical.

Equation (62) can be rewritten:

$$\frac{\kappa \mu \ell^2 \alpha_i^2}{1 - \alpha_i \ell} = \beta - \rho - \alpha_i \Lambda$$
(65)

For small  $\alpha_i$  the left-hand side of this equation is negligible, so:

$$\alpha_0 = (\beta - \rho) / \Lambda \tag{66}$$

Since

 $\alpha_0 \alpha_1 = \frac{\rho - \beta}{\ell \Lambda_c} \tag{67}$ 

the other  $\alpha$  is approximately

 $\alpha_1 \simeq \frac{\Lambda}{\ell \Lambda_c} \tag{68}$ 

Neglecting  $\alpha_0 \ell$  with respect to 1, this yields

$$a = \frac{2\alpha_0 \ell^2 \kappa \mu}{\Lambda} \tag{69}$$

If the reactor is close to critical

$$a = 2 \frac{\kappa \mu}{\beta} \quad (\alpha_0 \ell)^2 \tag{70}$$

Therefore, the disturbing effect is small when the coupling between core and reflector is weak, and when the reflector lifetime is short.

We try to estimate the effect for the experiment of Section 2. There

$$\alpha_0 \simeq 54 \text{ s}^{-1}$$

From (unpublished) pulsed neutron measurements in the same reactor we have found indications that

$$\frac{\kappa\mu}{\beta}=6; \qquad \ell=10^{-3} \text{ s}$$

Then:

a = 0.035

For the ratio of the two effects in (43) we then find

$$\frac{\frac{1}{2}Z_{1}^{2}f_{1}^{2}a}{\frac{1}{3}Z_{2}f_{2}} \approx \frac{0.070}{\beta} \approx 10$$

So the measured effect is about eleven times larger than the expected effect. This is in agreement with the experiment (about 14 times larger). So the modal effect indeed explains the observed discrepancy.

## 4. SUMMARY OF CONCLUSIONS

We summarize the main conclusions.

(i) When calculating  $\beta - \rho$  and  $\epsilon$  from threefold correlation experiments, one must take into account a space and energy correction factor.

This factor is expected not to deviate too far from 1. (ii) The moment

 $\overline{n(t) \{ n(t+\sigma) - n(t+\sigma+\tau) \}} n(t+2\sigma+\tau)$ 

is less sensitive to higher mode contamination than the moment with  $\sigma = 0$ .

- (iii) There are higher mode effects that cannot be removed by detector positioning. They depend on the system itself.
- (iv) From the results of the two simple calculations in Sections 3.6.2 and 3.6.3 we expect that the disturbance is small for a system consisting of a fairly reactive core, surrounded by a strongly absorbing blanket or reflector.
- (v) It is a difficult problem to evaluate the disturbance effect quantitatively for an actual fast system.

## ACKNOWLEDGEMENT

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## DISCUSSION

D. STEGEMANN: You state that the threefold correlation experiment is particularly suited to measuring  $\beta_{eff}$  in fast reactor systems without any other static experiments. In view of the relatively small detection efficiencies (about 10<sup>-5</sup>) in fast systems and seeing that  $Z_1$  is proportional to  $\epsilon$  and the threefold correlation term  $Z_2$  is proportional to  $\epsilon^2$  (i.e. about 10<sup>-10</sup>), I am almost sure that no reasonable results for  $\beta$  can be found using this method, especially as in addition one must distinguish between the two types of threefold correlation (which in Fig.1 you denote as "trees" a and b). Could you tell us why you believe this experiment to be more advantageous for measuring  $\beta_{eff}$  than a normal correlation noise experiment together with a static experiment for measuring the absolute reactor power?

J.B. DRAGT: With regard to your first point, I can say that here we have to distinguish between the "tree-b" effect, which is not proportional to Z, but to  $Z_1^2$ , and the "tree-a" effect, dependent on  $Z_2$ . Hence both effects are proportional to  $\epsilon^2$ , and the ratio of the two is independent of detector efficiency. This efficiency might give rise to problems in a fast reactor, although it is not clear to what extent this is a limitation, since detailed error evaluations for the third-order moments are not as yet available. If poor detector efficiency presents a problem, the number of detectors could be increased, in which case the problem is more of a practical than a fundamental nature.

As to your question, I do not believe that the usual correlation experiment, together with an absolute reactor power determination, is any more accurate than the present method, the advantage of which is that it provides a new and independent check on normal procedure. Moreover, it is interesting from the point of view of attempting to derive the maximum possible information from reactor noise.

G.S. BRUNSON: There is an interesting variation of the simple Rossi- $\alpha$  correlation experiment: One initiates the timing cycle with either a <sup>10</sup>B or <sup>235</sup>U counter, at positions of equal importance, and terminates the cycle in either case with a further counter at the centre of the core. On comparing the two correlation curves one finds that initiation with the <sup>235</sup>U counter gives a probability amplitude a few per cent larger than does the <sup>10</sup>B counter, due to the  $\nu$  neutrons introduced into the neutron population at time t = 0.

The fact that the additional neutrons produce an effect of only a few per cent is significant. Each neutron chain begins with a single fission, i.e. has an instantaneous population of  $\nu$ . However, the chains normally detected in the Rossi- $\alpha$  experiment by this comparison are shown to have a population of ~50 neutrons at  $t_{exp} = 0$ . It is clear then that the chains studied by this method are very much larger than average, having presumably experience some fortuitously large values of  $\nu$  before the first detection. The reason for this is quite clear, namely that a chain must be detected twice to be counted at all; hence chain detection probability is weighted as (pop.)<sup>2</sup>.

R.W. ALBRECHT: The fact that uranium and boron have different absorption cross-sections surely enters into account.

G.S. BRUNSON: The initiating counter merely detects the existence of a neutron chain and the sensitivity of the  $^{10}$ B or  $^{235}$ U counter is immaterial. What is important is to maintain exactly the characteristics of the terminating counter so that a precise comparison of the two experimental results may be made.
# ANALYSE DES TECHNIQUES DE MESURE DE LA REACTIVITE D'ENSEMBLES MULTIPLICATEURS FORTEMENT SOUS-CRITIQUES

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#### Abstract — Résumé

ANALYSIS OF THE TECHNIQUES FOR MEASURING THE REACTIVITY OF FAR SUB-CRITICAL MULTIPLYING SYSTEMS. The methods of measuring control-rod worth on the basis of reactor response to a periodic excitation source are analysed and it is shown that the ratio of the integrals of the prompt and delayed neutron densities over one period is independent of the shape of the signal used. This ratio will thus be equal to the ratio of the neutron densities in the presence of a time-independent source. The pulsed source, sinusoidal source and source removed methods therefore give identical worth values. Experimentation and numerical analysis show that the ratio of prompt to delayed neutron density, which is a linear combination of successive eigenvalues of the system, is largely dependent on the position of the source. It is demonstrated analytically that with a suitably chosen volume of integration, the contribution of the space harmonics is drastically reduced in the integrals of both the prompt neutron density and the delayed neutron density. Numerical studies show that the ratio of these two integrals is practically equal to the reactivity corresponding to the fundamental static mode and is almost independent of the position of the source. Integration of the neutron densities over a volume can be reduced to an integration along a straight line by a suitable choice of source location. Such an integration can be performed by means of an integral detector, thereby only requiring one measurement. Optimization of the position of the source and of the limits of integration to give the best value for the reactivity can be achieved on the basis of purely experimental criteria.

ANALYSE DES TECHNIQUES DE MESURE DE LA REACTIVITE D'ENSEMBLES MULTIPLICATEURS FORTEMENT SOUS-CRITIQUES. Les méthodes de mesure de l'antiréactivité à partir de la réponse d'un réacteur à une source d'excitation périodique sont analysées. Cette analyse montre que le rapport des intégrales sur une période des densités neutroniques prompte et retardée est indépendant de la forme du signal utilisé. Ce rapport sera par conséquent égal au rapport des densités neutroniques existant en présence d'une source constante. Les méthodes de la source pulsée, de la source sinusoïdale et de l'éjection de source conduisent donc à des valeurs identiques d'antiréactivité. Le rapport des densités neutroniques prompte et retardée, qui est une combinaison linéaire des valeurs propres successives du système, dépend fortement de la position: l'expérience et l'analyse numérique le montrent. On montre analytiquement que, lorsque le volume d'intégration est convenablement choisi, la contribution des harmoniques spatiales est fortement réduite tant dans l'intégrale de la densité neutronique prompte que dans l'intégrale de la densité retardée. Par une étude numérique, on montre que le rapport des deux intégrales est pratiquement égal à la réactivité correspondant au mode statique fondamental, et quasi indépendant de la position de la source. L'intégrale des densités neutroniques sur un volume peut se réduire à une intégrale simple par un choix judicieux de la position de la source: une telle intégration peut se réaliser au moyen d'un détecteur long et ne requiert qu'une seule mesure. L'optimisation de la position de la source et des limites d'intégration conduisant à la meilleure valeur de réactivité peut se faire sur base de critères purement expérimentaux.

#### 1. INTRODUCTION

C'est en raison du complet désaccord qui a été constaté entre les résultats théoriques et les résultats de mesure, en régime souscritique, de la reprise de réactivité des barres de contrôle dès que ces

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valeurs dépassent deux dollars, que ce travail a été entrepris. Pour expliquer ce désaccord, une des campagnes de mesure de l'expérience VENUS a été étudiée en détail.

La présente étude est essentiellement numérique, ses buts étant:

- de faire porter la comparaison entre la théorie et l'expérience sur des grandeurs directement mesurables et non sur les résultats indirects qui s'en déduisent, et de prouver ainsi la validité des résultats théoriques
- de prouver que, même dans des conditions défavorables, les résultats des méthodes de mesure proposées constituent une excellente approximation de la réactivité à mesurer.

L'intérêt de pouvoir mesurer correctement la réactivité ainsi que la constante de décroissance des neutrons prompts d'un ensemble fortement sous-critique est double. En effet, on peut

- mesurer la réactivité d'un réacteur toutes barres de contrôle enfoncées (caractéristique importante du point de vue de la sécurité)
- vérifier les méthodes théoriques sur la base de résultats expérimentaux obtenus sur des ensembles comportant de faibles quantités de combustible.

# 2. ANALYSE DES METHODES DE MESURE DE REACTIVITE EN REGIME SOUS-CRITIQUE

# 2.1. Définition de la réactivité statique d'un réacteur sous-critique

La réactivité statique est définie par la relation

$$\rho = \rho_1 = \nu_1 - 1$$

où

 $\nu_1$  représente la plus petite des valeurs propres de l'équation statique de Boltzmann

$$LN_n = \nu_n MN + RN_n$$

(1)

où

L = opérateur de destruction

M = opérateur de production

R = opérateur de transfert

N = densité neutronique.

La réactivité en dollars s'exprime par

$$\rho * = \frac{\rho_1}{\beta} = \frac{\nu_1 - 1}{\beta} = \frac{1 - k_1}{k_1 \beta}$$

( $\beta$  = fraction effective des neutrons retardés) puisque k<sub>1</sub>, le facteur effectif de multiplication, est égal à  $1/\nu_1$ .

La mesure de la réactivité  $\rho_1^*$  revient ainsi à mesurer la première valeur propre du système: il faut donc disposer de la fonction propre

correspondante, en l'occurrence de la distribution de la densité neutronique fondamentale.

D'une façon générale, la réactivité statique d'un réacteur souscritique sur le mode n est défini par

$$\rho_n = \nu_n - 1$$

où 
$$\rho_n^* = (\nu_n - 1)/\beta$$
, avec  $1 < \nu_1 \dots < \nu_{n \neq 1}$  et donc  $\rho_1 < \rho_{n \neq 1}$ .

# 2.2. Réponse d'un réacteur sous-critique à une source d'excitation variable dans le temps

En présence d'une source périodique de période T, suivant [1], le rapport des densités neutroniques promptes et retardées intégrées sur cette période vaut

$$\frac{A_{1}(\vec{r})}{A_{2}(\vec{r})} = \frac{\int_{0}^{T} N_{p}(\vec{r}, t) dt}{\int_{0}^{T} N_{r}(\vec{r}, t) dt} = \frac{\sum_{n=1}^{\infty} \frac{k_{n}}{1 - k_{n}(1 - \beta)} R_{n}(\vec{r}) \int_{0}^{T} S_{n}(t) dt}{\sum_{n=1}^{\infty} \frac{k_{n}}{1 - k_{n}(1 - \beta)} R_{n}(\vec{r}) \int_{0}^{T} \lambda C_{n}(t) dt}$$

où

 $N_p(\vec{r},t) = \text{densité neutronique totale prompte}$   $N_r(\vec{r},t) = \text{densité neutronique totale retardée}$  $avec N = N_p + N_r = \text{densité neutronique totale.}$ 

 $\sim$  N<sub>p</sub> est la densité neutronique qui existerait en l'absence de précurseurs de neutrons retardés, N<sub>r</sub> est la densité neutronique due aux seuls précurseurs des neutrons retardés.

Mais

$$k_n = \frac{1}{\nu_n}$$

et [1] donne

$$\int_{0}^{T} S_{n}(t) dt = (1 - k_{n}) K_{n} \int_{0}^{T} T_{n}(t) dt$$

$$\int_{0}^{T} \lambda C_{n}(t) dt = \beta k_{n} K_{n} \int_{0}^{T} T_{n}(t) dt$$

Nous obtenons après élimination de  $\int_{0}^{1} \lambda C_{n}(t) dt$ 

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$$\frac{A_{1}(\vec{r})}{A_{2}(\vec{r})} = \frac{\sum_{n=1}^{\infty} \frac{1}{\nu_{n} - (1 - \beta)} R_{n}(\vec{r}) \int_{0}^{T} S_{n}(t) dt}{\sum_{n=1}^{\infty} \frac{1}{\nu_{n} - (1 - \beta)} R_{n}(\vec{r}) \beta \frac{1}{(\nu_{n} - 1)} \int_{0}^{T} S_{n}(t) dt}$$
(2)

Dans cette expression,  $R_n(\vec{r})$  est la fonction propre d'ordre n et  $\nu_n$  la valeur propre correspondante de l'équation statique de Boltzmann appliquée au réacteur envisagé. Les fonctions  $S_n(t)$  sont les coefficients du développement en série de fonctions propres de  $S(\vec{r}, t)$ 

 $S(\vec{r}, t) = \Sigma S_n(t) R_n(\vec{r})$ 

En pratique, les différents modes sont excités de la même manière, de sorte que

$$S(\vec{r}, t) = \Sigma S_n(t) R_n(\vec{r}) = S(t)\Sigma S_n R_n(\vec{r})$$
 (3)

où les  $S_n$  représentent les coefficients du développement de  $S(\vec{r})$  en série de fonctions propres du problème.

Il vient donc

$$\frac{A_{1}(\vec{r})}{A_{2}(\vec{r})} = \frac{\sum_{n=1}^{\infty} \frac{1}{\nu_{n} - (1 - \beta)} S_{n}R_{n}(\vec{r})}{\sum_{n=1}^{\infty} \frac{1}{\nu_{n} - (1 - \beta)} S_{n}R_{n}(\vec{r}) \frac{\beta}{\nu_{n} - 1}}$$
(4)

 $A_1(\vec{r})/A_2(\vec{r})$  est donc indépendant de la forme du signal extérieur et en présence d'une source fixe

$$\frac{A_1(\vec{r})}{A_2(\vec{r})} = \frac{N_p(\vec{r})}{N_r(\vec{r})}$$

Les méthodes de Sjöstrand, de la modulation et de l'éjection de source fourniront donc des valeurs identiques pour le rapport  $A_1(\vec{r})/A_2(\vec{r})$ .

Posons, dans l'expression (4)

$$\frac{1}{\nu_n - (1 - \beta)} \frac{1}{\nu_n - 1} S_n = F_n$$

et

$$\nu_n - 1 = \rho_n$$

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(5)



FIG.1. Réponse d'un réacteur sous-critique à une source neutronique pulsée périodique.

Il vient

$$\frac{A_{1}(\vec{r})}{A_{2}(\vec{r})} = \sum_{n=1}^{\infty} F_{n} \rho_{n}^{*} R_{n}(\vec{r})$$

$$\sum_{n=1}^{\infty} F_{n} R_{n}(\vec{r})$$
(6)

Les  $F_n$  sont les coefficients du développement de  $A_2(\vec{r})$  en série de fonctions propres; les  $F_n \rho_n$  sont les coefficients du développement correspondant de  $A_1(\vec{r})$ .

Lorsque seul F<sub>1</sub> est différent de zéro,

$$\frac{A_1(\vec{r})}{A_n(\vec{r})} = \rho *$$

# 2.3. Aperçu des méthodes expérimentales

2.3.1. Méthode de la source pulsée périodique [1]

La réponse (la variation de la densité neutronique) en un point  $\vec{r}$  du réacteur est illustrée à la figure 1.

La période T doit être telle que

$$\frac{1}{\lambda_{\max}} \gg T \gg \Lambda$$

où  $\Lambda$  représente le temps de génération des neutrons prompts et  $\lambda_{max}$  la plus grande des constantes de décroissance des précurseurs des neutrons retardés.

Le rapport  $A_1/A_2 = \Sigma F_n \rho_n^* R_n(\vec{r})/\Sigma F_n R_n(\vec{r})$  vaut  $\rho^*$  lorsque seul  $F_1$  est différent de zéro, c'est-à-dire lorsque les neutrons sont distribués suivant la fondamentale.

2.3.2. Méthode d'extinction de source [2]

La réponse d'un réacteur sous-critique à une extinction de source est représentée à la figure 2. Lorsque la source fonctionne, la densité







FIG. 3. Réponse d'un réacteur sous-critique à une source sinusoidale.

neutronique vaut la somme des densités promptes  $N_p$  et retardées  $N_r$ . Par extrapolation, on détermine  $N_r$ . Le rapport  $N_p/N_r = \rho * \text{ si } N_p$  et  $N_r$  sont distribués suivant la fondamentale.

2.3.3. Méthode de la source modulée [3]

En présence d'une source modulée, soit

$$S = S_0 + \Delta S e^{j\omega t}$$

la réponse

$$N = N_0 + \Delta N e^{j(\omega t + \varphi)}$$

illustrée à la figure 3 permet la détermination du rapport  $A_1(\vec{r})/A_2(\vec{r})$ . Lorsque le taux de modulation  $\Delta S/S_0 = 1$  et lorsque la pulsation est optimale,

$$[\hat{\omega} = f(\lambda_i, \beta_i, \rho, \Lambda)]$$

$$\frac{A_{1}(\vec{r})}{A_{2}(\vec{r})} = \frac{\Delta N(\vec{r})}{N_{0}(\vec{r}) - \Delta N(\vec{r})}$$





#### 2.3.4. Méthode de la source crénelée périodique

Cette méthode se rattache aux méthodes des neutrons pulsés et de l'extinction de source. La réponse du réacteur est illustrée à la figure 4. La période T du signal doit être telle que (voir 2.3.1)

$$\frac{1}{\Lambda_{max}} \gg T \gg \Lambda$$
$$T - d \gg \Lambda$$
$$d \gg \Lambda$$

d = durée de fonctionnement de la source pendant une période  

$$\Lambda$$
 = temps de génération des neutrons prompts  
 $\lambda_{max}$  = la plus grande des constantes de décroissance des précurseurs  
des neutrons retardés

Dans ces conditions

$$\frac{A_1(\vec{r})}{A_2(\vec{r})} = \frac{N_P(\vec{r})}{N_R(\vec{r})} \frac{d}{T} = \frac{N_T(\vec{r}) - N_R(\vec{r})}{N_R(\vec{r})} \frac{d}{T}$$

Les autres méthodes présentent, pour la détermination de  $\rho$ \*, des désavantages par rapport à la méthode de la source en crénaux:

- Méthode de la source pulsée: pour une même valeur de S(t)dt, les corrections de temps mort à introduire lors du comptage seront plus
- grandes, car  $S(t)_{pulse} \gg S(t)_{crénaux}$ - Méthode d'éjection de source: outre le faible taux de comptage, elle nécessite le calcul de N<sub>r</sub> et comporte donc les incertitudes liées au modèle ponctuel et aux constantes ( $\beta_i$ ,  $\lambda_i$ )
- Méthode de la source modulée: elle nécessite le calcul de la pulsation optimale  $\hat{\omega} = f(\rho, \beta_i, \lambda_i, \Lambda)$  et comporte donc les incertitudes liées au modèle ponctuel et aux constantes  $\lambda_i$ ,  $\beta_i$  et  $\Lambda$ .

#### 2.4. Aspect temporel de la réponse à une source variable

Considérons le cas d'un signal en crénaux, répondant aux conditions spécifiées précédemment. Après extinction de la source, la réponse prompte peut s'écrire

$$N_{p}(\vec{r}, t) = \sum_{n=1}^{\infty} F_{n} \dot{\rho}_{n}^{*} R_{n}(\vec{r}) e^{-\alpha_{n} t}$$

avec

$$\alpha_n = \frac{\rho_n + \beta_n}{\Lambda_n}$$

Lorsque seul F<sub>1</sub> est différent de zéro,

$$N_{p}(\vec{r}, t) = F_{1} \rho_{1}^{*} R_{1}(\vec{r}) e^{-\alpha_{1} t}$$

avec

$$\alpha_1 = \frac{\rho_1 + \beta}{\Lambda}$$

ce qui se représente par une droite en coordonnées semi-logarithmiques (fig. 5).

#### 2.5. Conditions de validité des méthodes

Le rapport  $A_1(\vec{r})/A_2(\vec{r})$  ne sera égal à la réactivité statique  $\rho^* = \rho_1^*$ que si  $N_p(\vec{r}, t)$  et  $N_r(\vec{r}, t)$  sont répartis suivant la fondamentale  $R_1(\vec{r})$ . Pratiquement,  $N_p$  et  $N_r$  sont répartis suivant la fondamentale:

a) Lorsque  $\rho$  est faible, c'est-à-dire pour une situation proche de la criticité, car, dans ces conditions (voir 2.2),

$$\frac{1}{\nu_n - (1 - \beta)} \frac{1}{\nu_{n-1}} S_n = F_n$$

soit

$$\frac{1}{\rho_n + \beta} \frac{1}{\rho_n} \quad S_n = F_n$$

Mais,  $\rho_1$  étant faible et  $\rho_1 < \rho_{n \neq 1}$ 

 $F_{1} \rho_{1} \gg F_{n \neq 1} \rho_{n \neq 1}$  $F_{1} \gg F_{n \neq 1}$ 



FIG.5. Aspect temporel de la réponse neutronique prompte à une extinction de source.

de sorte que

$$\frac{\mathbf{A}_{1}(\vec{\mathbf{r}})}{\mathbf{A}_{2}(\vec{\mathbf{r}})} = \frac{\sum_{n=1}^{\infty} \mathbf{F}_{n} \mathbf{R}_{n} \boldsymbol{\rho}_{n}}{\sum_{n=1}^{\infty} \mathbf{F}_{n} \mathbf{R}_{n}} \simeq \boldsymbol{\rho}_{1}$$

b) Lorsque les neutrons de source (14 MeV), après ralentissement et diffusion, ont une répartition proche de  $R_1(\vec{r})$ . En effet, dans ce cas

$$S_1 \gg S_{n \neq 1}$$
 et  $F_1 \gg F_{n \neq 1}$ 

# 2.6. Elimination des harmoniques

# 2.6.1. Méthode de Gozani [4]

Nous n'avons pas analysé cette méthode en détail car, comme Gozani le précise, la méthode n'est valable que si les modes fondamentaux statiques et dynamiques (en décroissance exponentielle) sont identiques. Ce n'est pas le cas pour un réacteur réfléchi ou, de façon plus générale, pour un ensemble à plusieurs zones dont les caractéristiques d'absorption sont fort différentes [5].

# 2.6.2. Méthode d'«intégration pondérée» (IP)

Dans le cas d'un réacteur nu et homogène, dans les limites de validité de la théorie de la diffusion à un groupe les fonctions propres successives sont orthogonales, de sorte que

$$\int_{\mathbf{r}} \mathbf{R}_{n}(\mathbf{r}) \mathbf{R}_{m}(\mathbf{r}) d\vec{\mathbf{r}} = \mathbf{C}_{mn} \boldsymbol{\delta}_{m}^{n}$$

et le rapport

$$\begin{bmatrix} \underline{A_1} \\ \overline{A_2} \end{bmatrix}_{IP} = \frac{\int \limits_{\substack{\mathbf{r} \in \text{acteur}}} R_1(\vec{r}) \sum_{n=1}^{\infty} \rho_n^* F_n R_n(\vec{r}) d\vec{r}}{\int \limits_{\substack{\mathbf{r} \in \text{acteur}}} R_1(\vec{r}) \sum_{n=1}^{\infty} F_n R_n(\vec{r}) d\vec{r}}$$

peut s'écrire

$$\left[\frac{A_1}{A_2}\right]_{IP} = \frac{\sum_{n=1}^{\infty} \rho_n^* F_n \int_{reacteur} R_n(\vec{r}) R_1(\vec{r}) d\vec{r}}{\sum_{n=1}^{\infty} F_n \int_{reacteur} R_n(\vec{r}) R(\vec{r}) d\vec{r}} = \frac{\rho_1 F_1 \int R_1^2 dr}{F_1 \int R_1^2 dr} = \rho_1^* = \rho^*$$

En pratique, cette méthode serait lourde à appliquer car elle nécessiterait en tout point du réacteur la mesure de

$$A_{1}(\vec{r}) = \sum_{n=1}^{\infty} \rho_{n}^{*} F_{n} R_{n}(\vec{r})$$
$$A_{2}(\vec{r}) = \sum_{n=1}^{\infty} F_{n} R_{n}(\vec{r})$$

et le calcul de

$$N_1(\vec{r}) = R_1(\vec{r})$$

Comme nous le montrerons plus loin, pour une certaine position de la source l'intégration de volume peut en pratique se réduire à une intégrale simple.

Notons que l'intérêt de cette méthode est académique, car elle se limite aux réacteurs non réfléchis et nécessite la connaissance de la fondamentale.

2.6.3. Méthode d'«intégration simple» (IS)

Considérons un réacteur «plan infini», nu et homogène. Nous savons que

$$R_n(x) = \sin \frac{n \pi x}{a}$$

a étant l'épaisseur extrapolée du réacteur.

En mesurant, au moyen d'un détecteur intégral de longueur égale à 2a/3, le rapport

$$\begin{bmatrix} \underline{A_1} \\ \underline{A_2} \end{bmatrix}_{IS} = \frac{a/6}{5a/6} \sum_{n=1}^{\infty} \rho_n^* F_n R_n(x) dx$$
$$\int_{a/6} \sum_{n=1}^{\infty} F_n R_n(x) dx$$

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$$= \frac{F_1 \rho_1^* - \frac{F_5}{5} \rho_5^* - \frac{F_7}{7} \rho_7^* + \frac{F_{11}}{11} \rho_1^* + \frac{F_{13}}{13} \rho_{13}^* - \frac{F_{17}}{17} \rho_{17}^* \dots}{F_1 - \frac{F_5}{5} - \frac{F_7}{7} + \frac{F_{11}}{11} + \frac{F_{13}}{13} - \frac{F_{17}}{17} \dots}$$
$$= \frac{F_1 \rho_1^* + [E_{\rho}]_{15}}{F_1 + [E]_{15}}$$

on voit que les harmoniques d'ordre 2,3 et leurs multiples sont éliminées et que

 $\left[\frac{A_1}{A_2}\right]_{IS} \simeq \rho *_1$ 

 $\mathbf{si}$ 

$$[E_{\rho}]_{IS} \ll F_{1} \rho_{1}^{*}$$
 et  $[E]_{IS} \ll F_{1}$ 

Une première objection à cette méthode est que, dans le cas d'un réacteur <u>réfléchi</u>,  $R_n(x) = G_n(\sin n \pi x/a) + f_n(x)$ . Mais  $f_n(x)$ , qui traduit l'influence du réflecteur sur la distribution neutronique dans le noyau, est négligeable vis-à-vis de  $G_n(\sin n \pi x/a)$  dans la majorité des cas pour  $a/6 \le x \le 5a/6$ .

2.6.4. Méthode des deux détecteurs (DD)

Pour un réacteur plan, la réponse sommée de deux détecteurs placés en x = a/3 et x = 2a/3 s'écrit

$$\begin{bmatrix} \frac{A_1}{A_2} \end{bmatrix}_{DD} = \frac{A_1(a/3) + A_1(2a/3)}{A_2(a/3) + A_2(2a/3)}$$
$$= \frac{F_1\rho_1 * + F_5\rho *_5 - F_7\rho_7 * + F_{11}\rho *_{11} - F_{13}\rho *_{13} \dots}{F_1 + F_5 - F_7 + F_{11} - F_{13} \dots}$$
$$= \frac{F_1\rho_1 * + [E_\rho]_{DD}}{F_1 + [E]_{DD}}$$

et ce rapport sera proche de  $\rho_1^*$  lorsque

$$[E_{\rho}]_{DD} \ll F_{1} \rho_{1}^{*} \text{ et } [E]_{DD} \ll F_{1}$$

Par rapport à la méthode précédente, celle-ci présente l'inconvénient que les coefficients de  $\rho_5^*$ ,  $\rho_7^*$  ... ne comportent pas les facteurs 5, 7, 11 .. et qu'elle sera forcément moins précise; en d'autres termes sa limite d'applicabilité sera plus faible, la limite d'applicabilité étant la valeur maximale de  $\rho$  pour laquelle l'approximation est valable.



FIG. 6. Illustration des conditions pratiques de mesure.

d = distance source-noyau; h = hauteur physique; h' = hauteur extrapolée.

2.6.5. Aspect temporel de ces méthodes

Considérons la réponse prompte à un signal en crénaux. Après extinction de la source,

$$N_{p}(\vec{r}, t) = \sum_{n=1}^{n=\infty} F_{n} \rho_{n}^{*} R_{n}(\vec{r}) e^{-\alpha_{n} t}$$

Cette réponse devient, dans la méthode de l'intégration pondérée,

$$[N_{D}(t)]_{IP} = F_{1} \tilde{\rho}_{1} e^{-\alpha_{1} t}$$

dans la méthode de l'intégration simple, pour un réacteur plan infini,

$$[N_{p}(t)]_{IS} = F_{1} \rho_{1}^{*} e^{-\alpha_{1}t} - \frac{F_{5}}{5} \rho_{5}^{*} e^{-\alpha_{5}t} - \frac{F_{7}}{7} \rho_{7}^{*} e^{-\alpha_{7}t} \dots$$

dans la méthode des deux détecteurs, pour un réacteur plan infini,

$$[N_{p}(t)]_{DD} = F_{1} \rho_{1}^{*} e^{-\alpha_{1}t} + F_{5} \rho_{5}^{*} e^{-\alpha_{5}t} - F_{7} \rho_{7}^{*} e^{-\alpha_{7}t} \dots$$

Il en résulte que la réponse du détecteur intégral et la réponse sommée des deux détecteurs ne comporteront pratiquement plus que la décroissance des neutrons prompts suivant le mode fondamental.

# 2.7. Conditions pratiques de mesure

Le but poursuivi est l'élimination de l'influence des harmoniques sur la détermination expérimentale de  $\rho^* = \rho_1^*$  et la simplification maximale des mesures.

Considérons le cas le plus fréquent d'un réacteur cylindrique réfléchi. Si la source d'excitation est placée sur l'axe de ce réacteur (fig. 6) à une distance optimisée [6] de façon que la répartition radiale de la densité neutronique soit proche de la fondamentale, le rapport  $A_1(r,z)/A_2(r,z)$  ne sera plus fonction que de z, les harmoniques radiales étant absentes.

Cela simplifiera considérablement la mesure car,

- pour la méthode d'intégration simple, une mesure au moyen d'un compteur BF<sub>3</sub> (situé éventuellement dans le réflecteur radial) de longueur égale aux deux tiers de la hauteur extrapolée du réacteur suffira,
- pour la méthode des deux détecteurs, seule une mesure de la réponse sommée de deux détecteurs situés en z = h!/3 et z = 2h!/3 (h!: hauteur extrapolée) sera nécessaire.

Il va de soi que ces mesures devront être indépendantes de la position radiale des détecteurs.

Ces considérations présupposent que les distributions axiales de densité neutronique sont de la forme sin  $n \pi z/h^{1}$ . Si en pratique les distributions suivant z ne sont pas de cette forme (par exemple en raison de l'introduction partielle de barres absorbantes), il n'est plus possible de définir analytiquement les conditions favorables à l'élimination des harmoniques.

Il est cependant possible de définir expérimentalement les conditions optimales de mesure comme celles correspondant à  $\log[N_p(t)] = f(t)$ , après extinction de la source, se rapprochant le plus d'une droite.

Pour la méthode des deux détecteurs cette optimisation se résume à un déplacement de ces détecteurs.

Pour la méthode d'intégration simple, une modification de la longueur du détecteur est en principe nécessaire. En pratique cependant, l'optimisation peut se ramener au déplacement d'un détecteur de longueur fixe, étant donné la faible contribution, du point de vue du taux de comptage, de l'extrémité du détecteur qui est la plus éloignée de la source.

#### 3. VERIFICATION NUMERIQUE DES METHODES PROPOSEES

### 3.1. Introduction

L'étude numérique a été effectuée en rapport avec l'expérience critique VENUS (fig. 7). Cette situation est complexe en ce sens que



FIG.7. Vue schématique du réacteur VENUS.

le réacteur est réfléchi et essentiellement hétérogène, et les valeurs de réactivité fort élevées: la vérification numérique des méthodes proposées sera d'autant plus probante.

Le premier but poursuivi est le calcul des réponses locales des détecteurs  $A_1(\vec{r})/A_2(\vec{r})$  et de les comparer aux valeurs mesurées.

La vérification numérique des méthodes de mesure proposées consiste à comparer la valeur de la réactivité calculée par intégration des réponses locales à la réactivité statique calculée de la façon classique. Les calculs sont effectués au moyen du code THREX [7]. Ils consistent à déterminer, en présence d'une source fixe de 14 MeV, les distributions des densités neutroniques promptes et totales.

Dans la suite nous appellerons – improprement d'ailleurs – réactivité locale  $\rho_{*}^{*}(\vec{r})$  le rapport

$$\rho_1^*(\vec{\mathbf{r}}) = \frac{N_p(\vec{\mathbf{r}})}{N(\vec{\mathbf{r}}) - N_p(\vec{\mathbf{r}})} = \frac{N_p(\vec{\mathbf{r}})}{N_r(\vec{\mathbf{r}})}$$



FIG. 8. Distribution de  $\rho_1^{*}(\vec{r})$  en présence d'une source ponctuelle de 14 MeV située dans le réflecteur (avec quatre barres de contrôle).

## 3.2. Résultats

Plusieurs cas ont été envisagés, tant au point de vue du nombre de barres insérées qu'au point de vue de la distribution spatiale et énergétiqu de la source d'excitation.

3.2.1. Cas de quatre barres centrales insérées, la source d'excitation de 14 MeV étant logée dans le réflecteur (fig. 7)

La réactivité  $\rho = \nu_1 - 1 = 8,12\%$ , soit 11,04\$ ( $\beta_{eff} = 7,35 \cdot 10^{-3}$ ), constitue la valeur de référence. La distribution de  $\rho_1(\vec{r}) = A_p(\vec{r})/A_r(\vec{r})$ est donnée à la figure 8. On remarque la variation très importante de  $\rho_1(\vec{r})$  en fonction de  $\vec{r}$ . La valeur expérimentale mesurée dans le tube

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modérateur I.60 (fig. 7) vaut 6,83 \$, contre 6,22 \$ pour le calcul. L'accord est donc satisfaisant étant donné que le calcul se fait dans un plan et suppose a priori que la distribution axiale de densité neutronique correspond à la distribution fondamentale (introduction de  $B_{1H}^2$ ). L'allure de la variation de  $\rho_1(\vec{r})$  théorique a pu être comparée aux valeurs expérimentales. L'accord est excellent.

Pour la méthode d'intégration simple, l'aire d'intégration bidimensionnelle correspond au noyau proprement dit car les dimensions radiales du noyau correspondent aux deux tiers des dimensions radiales extrapolées. Si on compare la valeur trouvée

$$\left[\frac{A_1}{A_2}\right]_{IS} = \frac{\int N_p d\vec{r}}{\int N_r d\vec{r}} = 11,21$$

à  $\rho * = 11,04$  \$, on voit que l'accord est excellent. La méthode des deux détecteurs fournit

$$\left[\frac{A_1}{A_2}\right]_{DD} = 11,53$$

les deux détecteurs étant supposés être placés sur l'axe de symétrie de l'ensemble réacteur-source, à une distance du centre égale à 1/6 de la dimension extrapolée.

# 3.2.2. Cas de dix-huit barres enfoncées

De façon à définir les limites dans lesquelles  $[A_1/A_2]_{IS}$  et  $[A_1/A_2]_{DD}$ correspondent à  $\rho = \nu_1 - 1$ , ce cas extrême a été envisagé. Dans ces conditions, la valeur à mesurer est

$$\rho * = \frac{\nu_1 - 1}{\beta_c} = \frac{37.1 \cdot 10^{-2}}{7.35 \cdot 10^{-3}} = 50.48$$

Différentes conditions, tant au point de vue de la distribution spatiale qu'énergétique, de la source d'excitation ont été envisagées. Ces conditions ainsi que les résultats qui y correspondent sont donnés au tableau I.

# 3.3. Discussion des résultats

Le fait que, en présence d'une source d'excitation identique à la source de neutrons de fission du problème statique, la valeur de  $\rho_1(\vec{r})$  ne soit pas constante indique que la valeur de  $\beta$  à introduire dans la relation (4) doit être fonction de  $\vec{r}$  (fig.11). Cette modification de  $\beta$  est due à la modification du spectre rapide aux approches du réflecteur. L'augmentation de  $\rho_1(\vec{r})$  à l'approche du réflecteur s'observe expérimentalement.

Dans un travail ultérieur, nous envisageons de définir les valeurs locales de  $\beta$ ,  $\Lambda$ , etc., et de les relier aux valeurs moyennes classiques.

Source d'excitation	ρ <sub>1</sub> ( <b>r</b> )	$\begin{bmatrix} A_1 \\ A_2 \end{bmatrix}_{\text{IS}}$ (\$)	$ \begin{bmatrix} \frac{A_1}{A_2} \end{bmatrix}_{DD} $ (\$)
Ponctuelle, 14 MeV, dans le réflecteur	Fig.9	53,26	37,33
Ponctuelle, 14 MeV, au centre du noyau	Fig.10	44,25	53,9
14 MeV, uniformément répartie	-	50,5	-
Uniformément répartie, et distribuée en énergie suivant le spectre total de fission	-	49,8	-
14 MeV, distribuée suivant la source de fission du problème statique	-	50,8	49,33
Identique, tant du point de vue spatial que du point de vue énergétique, à la source de fission du problème statique	-	49,5	49,10

#### TABLEAU I. CONDITIONS ET RESULTATS

Cela se justifie d'autant plus pour des réacteurs à deux types de combustible (U, Pu).

Pour une distribution uniforme de la source, que son énergie soit de 14 MeV ou distribuée suivant le spectre total de fission, la valeur de  $[A_1/A_2]_{IS}$  est pratiquement égale à  $\rho * = \rho_1^*$ .

Lorsque la source d'excitation est située dans le réflecteur radial, la valeur de  $[A_1/A_2]_{IS}$  est proche de  $\rho_1^*$  mais la valeur de  $[A_1/A_2]_{DD}$  n'est plus satisfaisante.

Comme, pour une valeur de  $\rho_1^* = 50$  \$, les écarts entre  $\rho_1^*$  et  $[A_1/A_2]_{IS}$  sont au maximum égaux à 5 \$, et comme l'écart relatif augmente avec  $\rho_1^*$  (importance relative des harmoniques plus grande), l'erreur commise ne serait pour  $\rho_1^* = 30$  \$ que de 2 \$, et pour  $\rho_1^* = 10$  \$ de 0,2 \$. Cela se confirme, l'écart entre  $\rho_1^*$  et  $[A_1/A_2]_{IS}$  et  $[A_1/A_2]_{DD}$  étant très faible dans le cas de quatre barres insérées (3.2.1).

Ces précisions sont très satisfaisantes étant donné que l'incertitude sur la mesure expérimentale est de l'ordre de grandeur des écarts précités.



FIG.9. Distribution de  $\rho_1^*(\vec{r})$  en présence d'une source ponctuelle de 14 MeV située dans le réflecteur (avec dix-huit barres de contrôle).

#### 4. CONCLUSIONS

Deux méthodes de mesure de la réactivité  $\rho^* = \rho_1^*$  correspondant au mode fondamental sont proposées: la méthode d'intégration simple (2.6.3) et la méthode des deux détecteurs (2.6.4). Ces deux méthodes sont fondées sur l'élimination maximale de l'influence des harmoniques: cette élimination s'effectue selon un critère expérimental (2.7).

Par une analyse purement numérique, il a été montré qu'il est possible de mesurer des valeurs de réactivité jusqu'à 50 \$ avec une erreur inférieure à l'incertitude expérimentale, et cela dans le cas d'un réacteur complexe, par le fait qu'il est réfléchi, essentiellement hétérogène et perturbé par la présence de barres de contrôle.





FIG.11. Profil de  $\rho_1$  le long de l'axe de symétrie du réacteur, en présence d'une source d'excitation ayant la distribution fondamentale.

Il serait souhaitable que les méthodes proposées soient vérifiées expérimentalement par une comparaison systématique des valeurs de  $\rho_1(\vec{r})$  calculées et mesurées, et des valeurs calculées de  $\rho_1^*$  et expérimentales de  $[A_1/A_2]_{IS}$  et  $[A_1/A_2]_{DD}$  pour différents chargements d'un réacteur cylindrique réfléchi. Cette étude permettrait de définir les limites d'applicabilité pratique des méthodes préconisées.

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# A STOCHASTIC CALCULATION OF FAST REACTOR GENERATION TIMES

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## Abstract

A STOCHASTIC CALCULATION OF FAST REACTOR GENERATION TIMES. Multigroup calculations for fast reactors are basically static in character, based on a representation of the system in which 25 or fewer groups are used to describe 5 or 6 decades in energy. While most of the uncertainty in calculations (as exemplified by generation times which are consistently 10 to 20% too low) is due to errors in crosssections, errors may also arise in averaging over broad groups; and in the treatment of elastic scattering by means of computed elastic removal cross-sections. Also, the multigroup method does not permit convenient detailed study of the physics of slowing down in fast reactors.

In this paper a new approach to calculation of fast reactor temporal and integral properties is described. With this method the neutron slowing down process is treated approximately as a discrete time, discrete state Markov process. Starting with an initial condition of a pulse of neutrons distributed with a fission spectrum, the neutron density as a function of energy and time over a large number (>100) of energy 'states' or 'groups' is evolved at discrete time steps. From this discrete representation of the time-energy dependent neutron density one is able to calculate a finely resolved discrete approximation to the distribution of slowing down times to first generation fission. The first time moment of this function is the generation time. Convolution of this distribution yields a function proportional to the time dependent response of a counter after a burst of fission spectrum neutrons. It is this function which is sampled in the Rossi- $\alpha$  experiment. Compared to the usual multigroup calculations the method has the advantage of not being directly dependent on spectral or adjoint calculations, and of being able to account for elastic scattering very accurately. Further, it is a dynamic calculation which follows the evolution of the neutron slowing down process in detail, as opposed to the usual calculations based on static fluxes. The method is versatile and may be applied to fast spectral and criticality calculations, and the usual spectral indicators such as central core fission ratios. Example calculations using the discrete state Markov method have been performed in systems with simple geometry. These calculational results are compared to those of other methods.

#### 1. INTRODUCTION

The usual, multigroup approach to calculating fast reactor integral parameters is basically static in character. The multiplication constant, k, and the time constant,  $\alpha$ , (negative by convention) are essentially eigenvalues of the time separable multigroup equations. The prompt generation time is commonly evaluated indirectly from the relation [1],

$$\Lambda_{p} = \frac{\frac{p}{p}}{k_{\alpha}\alpha} = \frac{\frac{p}{p}}{k_{p}}$$
(1)

which clearly amplifies any errors in the value of the prompt multiplication constant,  $k_p$ , that is associated with  $\alpha$ .  $l_p$  is the prompt "lifetime" normally reported in the literature. Neutron slowing down is a continuous Markov process [2, 3], that is, a process for which the probability for being in a certain volume element of phase space (space-time-energy) at some future time depends only on the present state of the system and not on the past. In this paper we approximate the process as a discrete time, discrete state Markov process, and construct an array of state transition probabilities from simple probabilistic considerations of the collision physics. This matrix is then applied to evolve the time dependence of a pulse of neutrons on a discrete time-energy mesh. From this the usual integral parameters may be calculated directly but we obtain, in addition, a complete temporal description of the pulse. Space dependence is treated in the transport approximation.

# 2. REVIEW OF THE MATHEMATICS OF DISCRETE MARKOV PROCESSES

A discrete representation of the neutron density at any time,  $n\Delta t$ , is provided by the state vector [4],

$$\bar{\mathbf{s}}(\mathbf{n}\Delta t) = (\mathbf{s}_1, \mathbf{s}_2, \cdots, \mathbf{s}_i, \cdots, \mathbf{s}_N), \qquad (2)$$

where  $s_i$  is the probability that the neutron is in the state bounded by energies  $E_i$  and  $E_{i-1} (E_i < E_{i-1})$  at time  $n \Delta t$ . This presumes some initial state vector,  $\overline{s}_0$ , at zero time, which is in our case derived from the fission spectrum and for which the elements sum to unity.

Fundamental to the description of a discrete Markov process is the transition probability matrix,

 $\bar{\bar{P}}_{\Delta t} = \|P_{i, j}\|$ (3)

The element  $P_{i,j}$  is the probability that a neutron will make a transition from state i to state j in one time step,  $\Delta t$ . Clearly, if all possible states are included in the set we may write,

 $\sum_{j} \mathbf{P}_{i,j} = 1 \tag{4}$ 

The neutron state vector may be stepped forward in time using this matrix, thus,

$$\overline{s}(n\Delta t) = \overline{s}((n-1)\Delta t) \cdot \overline{\overline{P}}_{\Delta t} .$$
(5)

It is a well-known and particularly useful result of the theory of Markov processes that the probability of going from state i to state j in n time steps is given by the i, j<sup>th</sup> element of the n<sup>th</sup> power of the l-step matrix. Thus the n-step transition matrix is,

$$\bar{\bar{P}}_{\underline{n}\Delta t} = \bar{\bar{P}}_{\Delta t}^{\underline{n}} = \| P_{i,j}^{(n)} \|$$
(6)

The power is taken in the usual sense of matrix multiplication.

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# 3. A STOCHASTIC MODEL FOR SLOWING DOWN IN FAST SYSTEMS

The energy continuum is divided into N discrete states of which the first N-1 span the energy region of interest to fast reactors (say  $E_1 = 10$  MeV to  $E_{N-1} = 10$  eV). The first state is a  $\delta$ -function in energy. The N<sup>th</sup> state extends to zero energy and is an absorbing state for neutrons which survive to scatter below  $E_{N-1}$ . Its primary purpose is as a check to see that all neutrons are accounted for and Eq. (4) is satisfied. Three additional absorbing states are added for neutrons which suffer nonfission capture, leak from the system, or cause a fission. Thus the state vector has N+3 elements and the transition probability matrix is of rank N+3. At very long times the value of the N+1 st element of the state vector becomes the total probability for non-fission capture in the system. Similarly for elements N+2 and N+3.

For each state we define an expected time to collision in the state

$$\langle t \rangle = \gamma_i^{-1} = \left\langle \frac{1}{\nabla \Sigma_T(E)} \right\rangle$$
 (7)

The brackets imply an energy average over state i which will be considered shortly.

Now, the off-diagonal transition probabilities may be written as the product of two probabilities,

$$P_{i, j} = (1 - e^{-\gamma_i \Delta t}) P_{i, j}, \qquad i < j \le N$$
 (8)

The first factor is just the probability that a neutron suffers a collision during the time  $\Delta t$ . The second factor, called the transfer probability, is the probability that a neutron goes from state i to state j in a collision.

We must allow for the possibility that a neutron remains in the same state after a collision so that non-absorbing diagonal elements have the form

$$P_{i,i} = e^{-\gamma_i \Delta t} + (1 - e^{-\gamma_i \Delta t}) P_{i,i}, \quad i < N$$
 (9)

which is an accurate representation if  $\gamma_i \Delta t$  is small. The diagonal elements are unity for the four absorbing states,

$$\mathbf{P}_{i,i} = \mathbf{p}_{i,i} = 1$$
,  $i=N, N+1, N+2, N+3.$  (10)

Before defining the transfer probabilities we consider a method for computing expectations for a state (i.e. expected reaction rates, expected energy at which a collision occurs, etc.). Our fundamental assumption is that neutrons enter a state according to a distribution which is uniform in energy across the state. Thus neutrons about to suffer their first collision in state i are assumed to be distributed according to,

$$W_{1,i}(E) = \frac{1}{\Delta E_i} = \frac{1}{(E_{i-1} - E_i)}, \quad E_i \le E \le E_{i-1}$$
 (11)

If a large number of states is used this will be a good assumption for neutrons which enter the state via inelastic scattering or fission.

Elastic scattering is assumed isotropic in the center of mass system to make the above assumption hold. In regions of anisotropic scattering the scattering cross section is reduced by the factor  $(1 - \mu_c(E))$ 

$$\Sigma'_{2}(E) = (1 - \mu_{2}(E))\Sigma_{2}(E)$$
(12)

where  $\mu_c(E)$  is the average scattering cosine in the center of mass system, and the process is treated as being otherwise isotropic with scattered neutrons distributed according to the familiar relation

$$\frac{1}{(1-\alpha)E_{0}}, \qquad \alpha E_{0} \leq E \leq E_{0}$$
$$\alpha = \left(\frac{A-1}{A+1}\right)^{2}$$

where

and  $E_0$  is the energy at which scattering occurs. For each state we shall compute the appropriate average energy at which scattering occurs.

Should  $\alpha E_0$  lie between the state boundaries, the physics of the process is adjusted slightly so that the fundamental assumption holds. To do this we multiply the cross section by a scaling factor, a, which is close to unity, and at the same time divide the maximum energy loss per collision by a. The scaling factor is chosen so that the scattered neutron distribution extends just to the lower limit of a state. It can be shown that the moderating power of the isotope is preserved to high order if a is close to one and A > 10. If the scattered neutron distribution spans several states scaling is usually unnecessary.

Neutrons may survive to suffer more than one collision in a state so we define the  $n^{th}$  collision survival probability in state i,  $r_{i,n}$ , as the probability that a neutron survives to suffer at least n collisions in state i. Obviously,

$$r_{i,1} = 1$$
 (13)

If we assume that neutrons about to suffer their  $n^{th}$  collision in state i are distributed according to a function,  $w_{i,n}(E)$ , which is normalized to unity over the state, then we may define the normalized probability density for collision in state i

$$W_{i}(E) = \left[\sum_{n=1}^{\infty} r_{i,n} W_{i,n}(E)\right] / \left[\sum_{n=1}^{\infty} r_{i,n}\right]$$
(14)

If there is a collision in state i,  $W_i(E)dE$  is the probability that the

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collision occurs in dE about E (effectively time-averaged). Also,

$$\mathbf{r}_{i} = \sum_{n=1}^{\infty} \mathbf{r}_{i,n}$$
(15)

is the expected number of collisions a neutron will have while it is in state i.

 $W_i(E)$  is the meaningful weighting function for computing group expectations. Thus we might write the expected energy at which a fission occurs with isotope k in state i as

$$\langle \mathbf{E} \rangle_{i} = \frac{ \int_{\mathbf{E}_{i}}^{\mathbf{E}_{i}-1} \mathbf{E} \frac{\Sigma_{\mathbf{f},\mathbf{k}}(\mathbf{E})}{\Sigma_{\mathbf{T}}(\mathbf{E})} W_{i}(\mathbf{E}) d\mathbf{E} }{ \int_{\mathbf{E}_{i}}^{\mathbf{E}_{i}-1} \frac{\Sigma_{\mathbf{f},\mathbf{k}}(\mathbf{E})}{\sum_{\mathbf{T}_{i}}^{\mathbf{E}_{i}-1} W_{i}(\mathbf{E}) d\mathbf{E} }$$
(16)

or the expected time to collision in a state as

$$\gamma_{i}^{-1} = \left[ \int_{\mathbf{E}_{i}}^{\mathbf{E}_{i}-1} \frac{W_{i}(\mathbf{E})}{v\Sigma_{T}(\mathbf{E})} d\mathbf{E} \right]$$
(17)

The functions,  $w_{i,n}(E)$  and the parameters  $r_{i,n}$  are given by the relations

$$w_{i,n}(E) = \frac{r_{i,n-1}}{r_{i,n}} \sum_{k} \frac{a_{k}^{2}}{(1-\alpha_{k})} \int_{E}^{E} \frac{\Sigma'_{s,k}(E') w_{i,n-1}(E')}{\Sigma_{T}(E') E'}$$

and

$$r_{i,n} = r_{i,n-1} \sum_{k} \frac{a_{k}^{2}}{(1-\alpha_{k})} \int_{E_{i}}^{E_{i-1}} \int_{E}^{E_{i-1}} \frac{\sum_{s,k}'(E')w_{i,n-1}'(E')}{\Sigma_{T}(E')E'} dE' dE$$
(19)

which may be derived from the scattering physics with the scaling factor,  $a_k$ , for isotope k included. Of course, we assume that the lower limit of the scattered neutron distribution lies below  $E_i$  in deriving the last two equations.

The calculations presented in this paper assumed that cross sections were constant across a state. Some of the distribution and expectations for the constant cross section case are listed in Table I. Equivalent expressions for the first few collisions may be derived readily for resonance cross sections and cross sections whose energy variation across a state may be represented as  $E^{\lambda_i}$ ,  $\lambda_i$  any constant. With a fine enough state structure, most cases can be treated very accurately with these additional relations. Lack of space precludes their presentation here.

	TABLE I	
Dist	ributions and Expectations for Constant Cro	ss Sections <sup>a</sup>
β =	= $ln(\mathbf{E}_{i-1}/\mathbf{E}_i)$	
C =	$= \sum_{\mathbf{k}} \frac{\Sigma_{\mathbf{s},\mathbf{k}}(\mathbf{E})}{\Sigma_{\mathbf{T}}(\mathbf{E})} \frac{\mathbf{a}_{\mathbf{k}}^{2}}{(1-\alpha_{\mathbf{k}})}$	•
W <sub>i,n</sub> (E) =	$= \frac{C^{n-1}}{\Delta E_{i}r_{i,n}} \left[ \frac{\ell n^{(n-1)}(E_{i-1}/E)}{(n-1)!} \right] ,$	n = 1, 2, · · ·
r <sub>i,n</sub> =	$= C^{n-1}\left[1-\frac{1}{(\beta-1)}\sum_{j=1}^{n-1}\frac{\ell n^{j}\beta}{j!}\right],$	n = 2, 3, · · ·
$\langle E \rangle_i =$	$= \frac{\mathbf{E}_{i-1} + \mathbf{E}_{i}}{2\mathbf{r}_{i}} \sum_{\ell=1}^{\infty} \left(\frac{\mathbf{C}}{2}\right)^{\ell-1} \left[1 - \frac{1}{(\beta^{2}-1)} \sum_{j=1}^{\ell-1} \frac{(2\ell n\beta)^{j}}{j!}\right]$	
$\left\langle \frac{1}{E} \right\rangle_{i} =$	$= \frac{1}{r_i \Delta E_i} \sum_{\ell=1}^{\infty} \frac{C^{\ell-1} \ell n^{\ell} \beta}{\ell!}$	
$\langle v \rangle_i =$	$= \frac{2(\mathbf{v}_{i-1}^{2} + \mathbf{v}_{i-1}\mathbf{v}_{i} + \mathbf{v}_{i}^{2})}{3r_{i}(\mathbf{v}_{i-1} + \mathbf{v}_{i})} \sum_{\ell=1}^{\infty} \left(\frac{2C}{3}\right)^{\ell-1} \left[1 - \frac{1}{(\beta^{3/2} - 1)} \sum_{j=1}^{\ell-1} \frac{1}{j!}\right]$	$\left[ \left(\frac{3\ln\beta}{2}\right)^{j} \right]$
$\left\langle \frac{1}{v} \right\rangle_i =$	$= \frac{2}{\mathbf{r}_{i}(\mathbf{v}_{i-1}^{\dagger} + \mathbf{v}_{i})} \sum_{\ell=1}^{\infty} (2C_{i})^{(\ell-1)} \left[ 1 - \frac{1}{(\beta^{1/2} - 1)} \sum_{j=1}^{\ell-1} \frac{1}{j!} \right]$	$\left(\frac{\ell n\beta}{2}\right)^{j}$

<sup>a</sup>The expectations are presented here in the form of infinite series. In practice these series converge extremely rapidly and 3 to 5 terms are more than adequate.

The transfer probabilities follow directly from simple considerations of collision physics. We derive the diagonal elements of the transfer probability array and summarize the remaining elements.

If a neutron of energy E, in state i, has a collision, the probability that it remains in state i after the collision is given by a

summation over all the isotopes, of the probability that the collision is elastic and with isotope k,

$$(a_k \Sigma'_{s,k}(E)) / \Sigma_T(E)$$

times the probability that the final neutron energy is between  $\mbox{ E}$  and  $\mbox{ E}_{;}, \ \mbox{ i.e.,}$ 

$$\left(\frac{a_k}{1-\alpha_k}\right) \frac{(E-E_i)}{E}$$

for isotropic scattering (inelastic scattering within a state is insignificant).  $p_{i,i}$  is computed as the expectation over all collisions in state i,

$$p_{i,i} = \sum_{k} \frac{a_{k}^{2}}{(1-\alpha_{k})} \left\langle \frac{\Sigma'_{s,k}(E)}{\Sigma_{T}(E)} \left( \frac{E-E_{i}}{E} \right) \right\rangle_{i}, \quad i < N$$
(20)

For constant cross sections in state i we have,

$$\mathbf{p}_{i,i} = \left[1 - \mathbf{E}_{i} \left\langle \frac{1}{\mathbf{E}} \right\rangle_{i}\right] \sum_{k} \frac{\mathbf{a}_{k}^{2} \sum_{s,k}^{\prime}}{\sum_{T} (1 - \alpha_{k})}$$
(21)

 $Off\mbox{-}diagonal$  elements must include both elastic and inelastic scattering and are written

$$P_{i,j} = \sum_{k} \left[ \frac{a_{k}^{2}}{(1-\alpha_{k})} \left\langle \frac{\Sigma'_{g,k}(E)}{\Sigma_{T}(E)} + \frac{\Sigma_{j}(E)}{k} \right\rangle_{i} + \left\langle \frac{\Sigma_{ln,k}(E)}{\Sigma_{T}(E)} \right\rangle_{k} h_{k}(i \rightarrow j) \right],$$

$$\lim_{k \rightarrow j \leq N, \quad (22)} \frac{1 \leq i < N}{i < j \leq N, \quad (22)}$$

where

$$H_{i,j}(E) = \left\{ \begin{pmatrix} \frac{\Delta E_j}{E} & , \quad E_j \ge \left\langle \frac{1}{E} \right\rangle_i^{-1} \left( 1 - \frac{(1 - \alpha_k)}{a_k} \right) \\ \left[ \frac{E_{j-1}}{E} - \left( 1 - \frac{(1 - \alpha_k)}{a_k} \right) \right], \quad E_{j-1} > \left\langle \frac{1}{E} \right\rangle_i^{-1} \left( 1 - \frac{(1 - \alpha_k)}{a_k} \right) > E_j \quad (23)$$

$$0 \quad , \left\langle \frac{1}{E} \right\rangle_i^{-1} \left( 1 - \frac{(1 - \alpha_k)}{a_k} \right) \ge E_{j-1}$$

and the  $h(i \rightarrow j)$  are inelastic scattering transfer probabilities which must be estimated from available inelastic scattering data. Eq. (23) carries the implication that the appropriate average energy at which scattering occurs in a state is given by  $\langle \frac{1}{E} \rangle^{-1}$ . The remaining elements follow trivially:

$$p_{i,N+1} = \sum_{k} \left\langle \frac{\Sigma_{c,k}(E)}{\Sigma_{T}(E)} \right\rangle$$
(24)

$$P_{i, N+2} = \left\langle \frac{D(E)}{\Sigma_{T}(E)} \right\rangle B^{2}$$
(25)

$$P_{i, N+3} = \sum_{k} \left\langle \frac{\Sigma_{f, k}^{(E)}}{\Sigma_{T}^{(E)}} \right\rangle$$
(26)

Note that in this model leakage is treated in the transport approximation with an equivalent leakage cross section,  $D(E)B^2$ , included in the total cross section.

## 4. COMPUTATIONAL ASPECTS

Because fast reactor processes occur over a wide range of time scales (usually  $\gamma_1$  is of the order of 100 times greater than  $\gamma_{N-1}$ ) it is convenient to allow the time increment,  $\Delta t$ , to vary. This is done simply by applying Eq. (6). Typically, an array of transition probabilities is constructed which is based on a time step appropriate to the 1-10 Mev range of interactions. After a few iterations on the state vector the array might be squared twice to bring it to a time step of width  $4\Delta t$ . The process is repeated.

A similar procedure is used in building an array of transition probabilities. The first few rows are generated on a time,  $\delta t$ , such that

$$002 \ge \gamma_{i}\delta t \ge .001$$
,

with the remaining rows having only unit elements on the diagonal. This array is squared and new rows are added if the state collision rate per neutron satisfies

$$0.002 \ge \gamma_1(2\delta t) \ge 0.001$$
,

etc., to a value of n such that

$$2^{n}\delta t = \Delta t.$$

On a typical computer with a 32 K memory one may use a rank 125 transition probability array and have approximately half the core left for programming. Running times for a problem yielding all of the results described herein are of the order of two minutes for a 100 state description, essentially independent of the number of isotopes. This is obviously much less than would be required for a corresponding space independent Monte Carlo calculation.

#### 5. SPACE DEPENDENCE

One may readily generalize the discrete Markov slowing down model to space and angle dependence [5] however, except for very simple cases, computation becomes very complex and time consuming. The transition probability matrix must have two dimensions for each dimension that is added to the description.

One useful simple case is that of a fast reactor with a homogeneous core and a homogeneous reflector in spherical geometry. If the critical geometry is known, calculations of the temporal characteristics of a neutron pulse are carried out by assuming a suitable flux shape (perhaps based on experimental flux traverses) and coupling core and reflector with probabilities for transition between the two which are based on the diffusion theory derived currents at the interface. A four dimensional array is required for even such a simple calculation.

A program has been written and applied to the calculation of parameters in the spherical, reflected ZPR assembly 6F. A generation time of 54 ns was obtained at critical as compared to a value of 52 ns in the bare critical core [5].

# 6. APPLICATIONS

This model may be used to calculate most of the parameters normally obtained by multigroup techniques, but it is particularly well suited to the study of time dependent processes with prompt neutrons. All of the calculations presented here use cross sections derived from the ABN [6] 25 group set and have used 5 states for each of the first 20 ABN groups. It must be emphasized, however, that one of the main advantages of this model is its ability to represent cross sections and scattering physics in great detail. On conventional computers it is possible to repre-



FIG. 1. Time behaviour of a unit pulse in SUAK-1.

sent the system with as many as 60 states per decade in the range from 100 kev to 10 Mev, and 100 or more states per decade in the region below 100 kev.

A description of the behavior of a pulse of first generation prompt neutrons slowing down is obtained from the state vectors which, when each state value is divided by the lethargy width of the state, gives a discrete representation of the neutron density in lethargy. Figures 1 and 2 illustrate the evolution of a unit pulse in two representative systems, SUAK-1 [7], and the core of ZPR-III assembly 14 [1,8,9]. The dispersion is due mainly to inelastic scattering.



FIG.2. Time behaviour of a unit pulse in core of ZPR-III assembly 14.

Note that at any time more than 99% of the neutrons are distributed within 1 or 2 decades in energy. This fact may be used to extend the effective number of states greatly since at any time the transition matrix need only encompass the energies in which the neutrons exist.

A function of considerable interest is the distribution of slowing down times to first fission, defined in the continuum by [10],

$$K(t) = \int_{O} v_{p}(E) v \Sigma_{f}(E) N(E, t) dE$$
(27)

where N(E, t) is the first generation prompt neutron impulse response. If a pulse of unit area is used the zeroth time moment is the prompt multiplication constant,  $k_p$ . The first time moment of K(t) divided by kis the prompt lifetime,  $l_p$ , and the prompt generation time,  $\Lambda_p$ , is just  $p l_p/k_p$ . The Laplace transform of K(t) is related to the Laplace transform of the distribution for all fissions following the pulse, G(t), by [10, 11],

$$g(s) = \frac{k(s)}{1-k(s)} \quad . \tag{28}$$

G(t). is the function sampled in the Rossi- $\alpha$  experiment and may be computed directly from K(t) by numerical convolution.

The analog to Eq. (27) in our model is given by,

$$K(n\Delta t + \frac{\Delta t}{2}) = \sum_{j=1}^{N-1} s_j \sigma', P_{j,N+3}$$
(29)

where  $v'_{p,j}$  is an average prompt fission yield in the state and  $s_j$  is the j<sup>th</sup> element of the state vector at time  $n\Delta t$ . This function is plotted in Fig. 3 for four different fast systems, of which all but SUAK-2 are critical. The effect of the U-238 fission threshold is clearly visible in the curve for SUAK-1. Table II compares some of our generation time and lifetime calculations with other calculations and experiments.





Analogous distributions to leakage, L(t), capture, R(t), and neutron death in the system, T(t), may be calculated. For example,

$$L(n\Delta t + \frac{\Delta t}{2}) = s_{N+2}((n+1)\Delta t) - s_{N+2}(n\Delta t)$$
(30)

These functions are illustrated in Figs. 4 and 5 for two very different systems. Their moments are compared in Table II.

#### 7. CONCLUSIONS

We have presented an approach to fast reactor neutronics calculations which is based on a direct, time dependent, mathematical analog to a slowing down pulse of neutrons. In particular, we are able to evaluate generation times directly as the first time moment of the distribution to first fissions. Elastic scattering is treated by a more straightforward approach than the elastic removal cross sections normally used in multigroup theory. Our calculations have used a maximum of 100 energy states, but one may potentially use 300 or more states. Such a large number is more than adequate to detail the current knowledge of cross sections and scattering.

#### TABLE II

	Pu-239		Core ZPR3	Core ZPR 3
SYSTEM	Sphere	SUAK-1	Assy. 6F	Assy14
Composition,				
Atoms/cc x $10^{-22}$				
Carbon				5.350
Aluminum		0.437	1.900	
Stainless Steel			1.050	0.790
U-235		0.821	0.672	0.450
U-238		3.240	0.763	0.034
Pu-239	4.960			
Effective Multiplication				
Constant, k <sub>eff</sub>	1.00	0.858	1.00	1.00
Geometric Buckling				
$(cm^{-2})$	0.15400	0.01877	0.00704	0.00755
Central Core Flux				
Averaged Energy (Mev)	1.57	0.85	0.92	0.86
Bare Core Prompt Generation				
Time, $\Lambda_{\mathbf{p}}$ , (ns)	3.38	32.6	52.0	156.0
Bare Core Prompt Lifetime,				
l <sub>p</sub> , (ns)	3.37	27.7	51.6	154.9
Calculated Prompt Lifetime				
(1963 X-sections)			45.0 <sup>b</sup>	144.0 <sup>b</sup>
Calculated Prompt Lifetime				
(1966 X-sections)		27.25 <sup>c</sup>	68 <sup>d</sup>	153d
Experimental Prompt				
Lifetime (ns)		28.3 <sup>c</sup>	77 <sup>d</sup>	187d
Mean Time to Non-fission				
Capture (ns)	7.35	47.2	83.6	268.6
Mean Time to				
Leakage (ns)	2.84	22.4	34.0	44.2
Mean Time to Death by				
Leakage or Absorption	3.14	28.6	47.1	115.3
Percentage of Neutrons				
Lost by Capture	1.8	15.9	11.1	11.9
Percentage of Neutrons				
Lost by Leakage	65.0	50.7	49.5	48.0
Percentage of Neutrons				
Causing Fission	32.2	33.4	39.4	40.1

Properties of Four Fast Assemblies<sup>a</sup>

<sup>a</sup>Unless otherwise noted, figures are for bare critical cores and have been computed with the method of this paper.

<sup>b</sup>Reflected system, data of Davey, 1963 [8].

<sup>C</sup>Personal communication, E. Wattecamps of Kernforschungszenstrum Karlsruhe, September, 1967.

<sup>d</sup>Reflected system, data of Meneghetti and White, 1966 [1].

The use of the transport approximation is known to be sufficient for many fast systems [12]. Multigroup discrete ordinates calculations of k seldom differ by more than 2% from values obtained in the transport approximation in simple systems. This is well within the range for which k may be reliably estimated on systems for which no integral experimental data is available. Certainly any errors induced in the generation time and other temporal characteristics are far less than existing errors due to our ignorance of cross sections. Computing times are substantially less than those required by an alternate method, Monte Carlo, and the results are free of the statistical fluctuations inherent to Monte Carlo.



FIG.4. Time distributions in a bare, critical <sup>239</sup>Pu sphere.



FIG.5. Time distributions in core of ZPR-III assembly 14.

Certainly the discrete Markov model will not replace the time honored multigroup techniques, but it is expected to provide useful supplementary information about the physics of slowing down in fast reactors

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## DISCUSSION

B.I. SPINRAD: In the calculations you performed on reflected systems, how did you treat reflector events? Did you consider neutrons absorbed in the reflector as absorption, capture or leakage?

R.W. ALBRECHT: The calculations reported here were on equivalent bare systems and the reflector effect was corrected for by introduction of an appropriate term. In other calculations, not mentioned here, we used a two-region model with transitions between core and reflector calculated by including artificial transition elements in the Markov transition matrix.

B.I. SPINRAD: I see, but if the calculations were on equivalent bare systems augmented by reflector savings then it is not surprising that they did not agree very well with the experiments. R.W. ALBRECHT: Certainly. The experimental results were adjusted by Davey, who used some core reflector model to get an equivalent bare core lifetime. The equivalent bare core results, as calculated from the experimental measurements on this basis, may well be suspect.

B.I. SPINRAD: I should indeed like to commend the fact that in your paper the kinetic parameters obtained are well defined. The difficulties experienced in correlating theory and experiment are due, I feel, to the fact that we do not know yet what the experiments mean!

D. STEGEMANN: Igather from your paper, Mr. Albrecht, that timeindependent spectra, too, can be calculated fairly easily. Is this correct?

R.W. ALBRECHT: Yes, the time-independent spectrum can be directly calculated from the evolution of a generation by integrating N(E, t) over time. This has been done and is one option of the code.

D. STEGEMANN: Was the 14-MeV source spectrum of the neutron generator considered or was a fission neutron spectrum taken instead?

R.W. ALBRECHT: I think this point needs some clarification. Our calculation is <u>not</u> a simulated pulse neutron experiment, but rather a direct computation of the evolution of a neutron generation in a multiplying medium. Thus no external source enters the calculation. However, we have also used this method to calculate slowing-down parameters of moderators and, in such a case, a 14-MeV source is included; results then show such interesting effects as Plazek wiggles in the first few collisions.

H. RIEF: What is the initial neutron distribution function in space and energy for calculating eigenvalues? Did you iterate this distribution before making a fundamental-mode distribution and what are the criteria for this?

R.W. ALBRECHT: The initial neutron distribution in energy is, of course, the fission spectrum. In space, the distribution is essentially fundamental-mode although this does not have any direct effect since leakage is only calculated from an initial buckling value. Eigenvalues are not calculated since no iteration is done; instead, the reported parameters are calculated directly from the input constants with the multipli-

cation constant given by  $k = \int K(t) dt$ .
# INFLUENCE OF TIME-DEPENDENT COUPLING COEFFICIENTS AND DELAY TIMES ON THE KINETICS OF A MODULAR FAST REACTOR CORE\*

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### Abstract

INFLUENCE OF TIME-DEPENDENT COUPLING COEFFICIENTS AND DELAY TIMES ON THE KINETICS OF A MODULAR FAST REACTOR CORE. Previous treatments of kinetics problems in modular and toroidal cores in fast breeder reactors have been based on Avery's coupled kinetics equations for partial integral fission sources. Most of these calculations do not take account of the time dependence of the reactivity coupling coefficients caused by possible flux tilting in an excursion. Furthermore, the effect of time delays for neutrons travelling between the modules, which is expected to be small, is not included in the model itself. The present investigation of the behaviour of two coupled modules is based on our recent derivation of coupled reactor kinetics equations from the Boltzmann equation. The distribution functions of delay times for neutrons are approximated by delta functions. It is shown that in three-group theory an analytical expression for the average delay time between two modules can still be derived. The calculation is based on the three-group leakage fluxes. Cross-sections are condensed from a 26-group set. Step changes in fission cross-sections are considered as perturbations. Coupling coefficients are calculated using the critical flux distribution and, to show the effect of time dependence, using the asymptotic flux distribution. Finally, a numerical method for solving coupled reactor kinetics equations is outlined.

Comparing solutions of simple kinetics problems without feed-back but with time-dependent coupling coefficients and delay times to solutions without consideration of these two effects, it is found that the influence of neutron delay times is indeed negligible for the usual transients considered in the safety analysis of fast breeder reactors. However, the inclusion of time-dependent coupling coefficients can markedly alter the module power during an excursion.

### 1. INTRODUCTION

### 1.1. Modular core concepts

To overcome problems with the positive sodium void coefficient for large fast breeder reactor cores, modular [1], [2], and annular [3], core designs have been proposed. The modular design, consisting of Pu bearing modules surrounded by  $U^{2\,3\,8}$  breeding blanket regions which are separated from adjacent modules by thin graphite regions allows for building up cores of different sizes using varying numbers of these basic modules. The sodium void coefficient of each module is negative.

Other modular concepts proposed in the past are the molten Plutonium design [4] and, in a quite different connection, modular nuclear rocket designs [5].

<sup>\*</sup> This work was sponsored by the Texas A & M University Research Council.

### 1.2. Previous work and scope of this paper

Previous work on modular fast reactor kinetics and dynamics has been based on AVERY's work in connection with the coupled fast-thermal ZPR III experiments [6]. This model does not include finite delay times for neutron exchange between modules since their influence is expected to be small. It will be shown in this paper that for normal operating behavior and for the most likely type of excursions the delay times can indeed be neglected.

A more important question that has not been investigated in previous work [7], [8] is the possible time dependence of the coupling coefficients caused by a changing flux shape in the system and the influence of these time dependent coupling coefficients on the temporal behavior of the system.

By considering step changes in cross-sections of a system consisting of two modules we will show that the time dependence of coupling coefficients must be taken into account in the kinetics treatment of modular fast cores.

Finally we will outline a numerical solution method for kinetics problems in modular cores.

### 2. CALCULATIONAL MODEL

#### 2.1. Kinetics equations

Kinetics equations for a system of M coupled modules as derived from the Boltzmann equation by PLAZA [9], [10] will be used in this paper:

$$\frac{dT_{j}}{dt^{d}} = \frac{\rho_{j} + \gamma_{j} - \beta_{j}}{\Lambda_{j}} T_{j} + \sum_{i=1}^{N} \lambda_{ji} C_{ji} + \sum_{k=1}^{M} \sum_{\Lambda_{j}}^{\varepsilon} C_{ki} \int_{0}^{\infty} T_{k} (t-T) p_{kj} (T) dT$$
(1)
$$\frac{dC_{ji}}{dt^{ji}} = \frac{\beta_{j}}{\Lambda_{j}} T_{j} - \lambda_{ji} C_{ji}$$
(2)

where

 $T_{i}(t)$  amplitude function of neutron flux in module j  $C_{i}(t)$  " " of delayed neutron precursors. ji(t) " of delayed neutron precursors, module j, group i Λj prompt neutron generation time in module j effective delayed neutron fraction in module j λji delayed neutron decay constant, module j, group i effective reactivity of j-th isolated module externally impressed reactivity in module j coupling coefficient for neutron exchange k to j γj ρj εĸj p<sub>kj</sub> (T)dT distribution function of delay times for neutrons going from module k to module j (assumed to be time independent)

We note, that when the system is critical, all  $\gamma_j$  are negative. The reactivities, prompt neutron lifetimes and the coupling coefficients are rigorously defined in terms of the

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system's time dependent flux shape function. In the last term of Eq.(1) the time dependence of the prompt neutron generation time has been assumed to be negligible.

The distribution functions of delay times will be assumed to be delta functions:  $p_{kj}(T)dT = \delta(T - T_{kj})dT$ , where  $T_{kj}$  is a properly calculated average delay time. (For short time initial transients, as for instance in pulsed neutron experiments with coupled modules, more detailed descriptions of  $p_{kj}(T)dT$  may be required; we refer the reader to the work of ADLER, GAGE and HOPKINS [11].)

With the above substitution Eq. (1) reads:

$$\frac{\mathrm{d}T_{j}}{\mathrm{d}t^{*}} = \frac{\rho_{j} + \gamma_{j} - \beta_{j}}{\Lambda_{j}} T_{j} + \sum_{i=1}^{N} \lambda_{ji} C_{ji} + \sum_{k=1}^{M} \frac{\varepsilon_{kj}}{\Lambda_{j}} T_{k}(t - T_{kj})$$
(3)

At criticality,  $\rho_1 = 0$ , and we obtain the criticality condition of the system:

γ <sub>10</sub> +ε <sub>110</sub>	ε <sub>210</sub>	• • •	<sup>ε</sup> M10		
<sup>ε</sup> 120	-		•		
-	•	Yjo <sup>+€</sup> jjo	-	= 0	(4)
•			- •		
ε <sub>lMo</sub>	•	· · ·	γ <sub>Mo</sub> +ε <sub>MMo</sub>		

where the subscript zero stands for evaluation of the quantity at criticality. In the special case of two symmetrical modules:  $\varepsilon_{110} = \varepsilon_{220} = \varepsilon_R$  as well as  $\gamma_{10} = \gamma_{20} = \gamma_0$  and  $\varepsilon_{120} = \varepsilon_{210} = \varepsilon_T$ the criticality condition reads:

 $\gamma_{0} + \varepsilon_{R} + \varepsilon_{T} = 0$ 

We wish to call the j-th module strongly coupled if max  $\varepsilon_{kj} \gg \beta_j$ ; in this case the module would not retain its  $k \neq j$  isolated flux shape in the system. Modular fast breeder reactors are of this type.

Module j will be called weakly coupled if max  $\epsilon_{kj} \ll \beta_j$ ; here the module would approximately retain its isolated flux shape in the system.

### 2.2. Model geometry and composition

Case I: We consider a system of two initially symmetrical modules, 64 cm wide, separated by a 32 cm wide breeding blanket and enclosed by 32 cm breeding blankets. The blanket will only be considered at the beginning of life without Pu buildup.

The module and blanket material volume percentages, intended to be only roughly representative of an actual modular

(5)

system, are as follows; module: 50% Na, 19.6% Fe, 30.4%  $UO_2$ -PuO<sub>2</sub> of 90% theoretical density, and for the blanket: 20% C, 32% Na, 16% Fe, 32%  $U^{238}$ . The above percent composition of the fuel is: 79.52% $U^{238}$ , 12.90%Pu<sup>239</sup>, 6.14%Pu<sup>240</sup>, 1.02%Pu<sup>241</sup> and 0.41%Pu<sup>242</sup>.

Case II: The composition stays as in Case I, however the outside blankets are deleted. To demonstrate effects of weak coupling, the diffusion coefficients have been artificially reduced by a factor 70 in the core modules to reduce the leakage and with it the coupling. The width of the modules is only 16 cm in this case.

### 2.3. Calculation of coupling parameters

The calculation of the coupling parameters is performed within a one-dimensional, plane geometry, 3-group diffusion theory model. The lower energy limits of the groups are 0.8Mev, 0.1kev and 0.025ev. Cross-sections for the calculation were obtained by condensation of the 26 group set by BONDARENKO et al [12].



FIG.1. Power density; ----- critical system, --- 1% change in  $\nu \Sigma_{f}$ .

The steady state coupling coefficients  $\gamma_0$ ,  $\varepsilon_T$  and  $\varepsilon_R$ are quite difficult to evaluate from their formal definitions [10]. We will use reasonable approximations for them here that satisfy the criticality condition, based on eigenvalues of two multigroup diffusion calculations. The steady state isolated module reactivity is obtained from the eigenvalue  $k_0$  of a multigroup eigenvalue calculation for the isolated module as  $\gamma_0 = 1-k_0^{-1}$ . To calculate the steady state coupling parameters  $\varepsilon_T$  and  $\varepsilon_R$  the criticality condition for two symmetrical modules is used:  $\gamma_1 + \varepsilon_T + \varepsilon_R = 0$ . A fictitious multigroup eigenvalue calculation for the system is performed setting  $v\Sigma_f = 0$  everywhere except in one module. The resulting eigenvalue  $k_f$  leads to a negative reactivity  $\rho_f = 1-k_f^{-1}$ that must exactly be compensated for by the suppressed fissions; thus:  $\rho_f = -\varepsilon_T$ . (The relatively small contribution of U<sup>230</sup> contribution from the other core in this approximation). Knowing  $\gamma_0$  and  $\epsilon_T$ , we calculate  $\epsilon_R$  from the criticality condition.

The perturbation considered in case I will be a 1% step increase in  $\nu\Sigma_{\rm f}$  in module 1 and a simultaneous 1% decrease in  $\nu\Sigma_{\rm f}$  in module 2. The resulting tilt in power density P, as obtained from a  $\lambda$ -mode calculation, is shown in Fig. 1.

The  $\lambda$ -mode fluxes in group g,  $\psi^g$ , are a good approximation to the asymptotic flux shape for the perturbation considered and are used to calculate the asymptotic, or effective coupling coefficients that should be used in a rigorous kinetics calculation. Thus we have:

$$\gamma_{j} = \gamma_{j0} \frac{\int_{j} \sum_{m} \chi^{m} \phi_{j0}^{m+} \sum_{g} \nu \Sigma_{fj0}^{g} \psi^{g} dx}{\int_{j} \sum_{m} \chi^{m} \phi_{j0}^{m+} \sum_{g} \nu \Sigma_{fj}^{g} \psi^{g} dx}$$
(6)

$$\varepsilon_{kj} = \varepsilon_{T} \frac{\int_{k} \Sigma \chi^{m} \phi_{k0}^{m+} \Sigma v \Sigma_{fk}^{g} \psi^{g} dx}{\int_{j} \Sigma \chi^{m} \phi_{j0}^{m+} \Sigma v \Sigma_{fj}^{g} \psi^{g} dx}$$
(7)

$$\rho_{j} = \frac{\int_{j} \sum_{m} \chi^{m} \phi_{j0}^{m+} \sum_{g} \Delta(\nu \Sigma_{fj}^{g}) \psi^{g} dx}{\int_{j} \sum_{m} \chi^{m} \phi_{j0}^{m+} \sum_{g} \nu \Sigma_{fj}^{g} \psi^{g} dx}$$
(8)

In equations (6) through (8)  $f_{,}$  dx means integration over the j-th module and  $\phi_{jo}^{m+}$  is the adjoint flux in group g of the j-th module when isolated from the critical system.

If  $v\Sigma_{fl} > v\Sigma_{fl0}$  and  $v\Sigma_{f2} = v\Sigma_{f20}$ , then we see from equation (7) that  $\varepsilon_{21} < \varepsilon_T$ , meaning that neutrons supplied from module 2 to module 1 now contribute less equivalent reactivity in module 1. The coupling coefficients  $\varepsilon_{kj}$  are to be viewed in the sense of equivalent reactivities and not in terms of expectation values for causing fissions.

The reflection coupling coefficients,  $\epsilon_{\rm R}$ , are assumed to be unchanged by the particular perturbation considered since most reflected neutrons return from the blankets after travelling only a few mean free path and the blankets are not perturbed.

The calculations for Case II are analogous to Case I except that the perturbations of the fission cross section are smaller; in module 1: 0.5% increase in  $\nu \Sigma_{f}$  and in module 2: 0.5% decrease in  $\nu \Sigma_{f}$ .

Results of these calculations are summarized in TABLE I, where we note that generally  $\varepsilon_{\rm T}\ll\varepsilon_{\rm R}$  and that in Case I:  $\varepsilon_{\rm T}$  = 5.65 x  $\beta({\rm strong\ coupling})$  whereas in the fictitious Case II:  $\varepsilon_{\rm T}$  = 0.0041 x  $\beta({\rm weak\ coupling})$ .

### TABLE I

Summary of	input	data
------------	-------	------

	Case I		Cas (weak d	Case II (weak coupling)		
	Module l	1% change νΣ	f Module 1	0.5% change $v\Sigma_{f}$		
	Module 2	-1% "	" Module 2	-0.5% ""		
	critical	asymptotic	critical	asymptotic		
Y,	-3.9020/-1 <sup>a</sup>	-3.8634/-1	-2.8304/-3	-2.8163/-3		
Ŷ	-3.9020/-1	-3.9414/-1	-2.8304/-3	-2.8446/-3		
ε <sub>R</sub>	3.5971/ <b>-</b> 1	3.5971/ <b>-</b> 1	2.808/-3	2.808/-3		
ε	3.049/-2	3.6358/-2	2.240/-5	3.7341/-5		
ε	3.049/-2	2.5569/-2	2.240/-5	1.3437/-5		
ρ	0	9.9010/-3	0	4.9751/-3		
ρ	0	-1.0101/-2	0 .	-5.0251/-3		
٨	2.193/-7	2.4338/-7	7.923/-8	7.9305/-8		
٨	2.193/-7	2.4918/-7	7.923/-8	8.0103/-8		
β <sup>2</sup>	5.40/-3	5.40/-3	5.40/-3	5.40/-3		
Τ <sub>m</sub>	2.0471/-7	2.0471/-7	6.990/-8	6.990/-8		
T <sub>R</sub>	4.6381/-8	4.6381/-8	2.500/-8	2.500/-8		
			5			

a/-x means  $10^{-x}$ 

The average delay times  ${\rm T}_{\rm R}$  and  ${\rm T}_{\rm R}$  for transmission and reflection respectively are calculated from the respective 3 group delay times by

 $T_{T} = \frac{\sum_{z} j_{g}^{+}(b, \infty) T_{T}^{g}}{\sum_{z} j_{g}^{+}(b, \infty)}$  $T_{R} = \frac{\sum_{z} j_{g}^{-}(o, \infty) T_{R}^{g}}{\sum_{z} j_{g}^{-}(o, \infty)}$ (10)

where we consider a one dimensional coupling slab  $0 \le x \le b$ , that is illuminated at  $t = o_1$  and x = o with a source of neutrons having the same leakage spectrum as one of the core modules. The other face at x = b is bounded by a black absorber,

(9)

such that  $\phi(b,t) = 0$ .  $j_{+}^{+}(x,t)$  and  $j_{-}^{-}(x,t)$  are the partial group currents in the positive and negative x-direction respectively.

We first define the time integrated transmission neutron group currents at x = b

$$Q_{\mathbf{f}}^{\mathbf{f}}(\mathbf{b},\mathbf{t}) = \int_{\mathbf{c}}^{\mathbf{t}} \mathbf{j}_{\mathbf{g}}^{\mathbf{t}}(\mathbf{b},\mathbf{t}')d\mathbf{t}' \qquad (11)$$

and the time integrated reflection neutron group currents at  $\boldsymbol{x}$  =  $\boldsymbol{o}$ 

$$Q_{\rm R}^{\rm g}(o,t) = \int_{\rm c}^{\rm t} j_{\rm g}^{-}(o,t')dt' \qquad (12)$$

The combined boundary - initial value problem

$$(\frac{1}{v})^{g} \frac{\partial}{\partial t} \frac{\phi^{g}(x,t)}{b} = D^{g} \frac{\partial^{2} \phi^{g}(x,t)}{\partial x^{2}} - \Sigma^{g} \phi^{g}(x,t) + \Sigma \Sigma^{h+g} \phi^{h}(x,t)$$

$$\phi^{g}(b,t) = o \quad (t \ge o)$$

$$\phi^{g}(x,o) = o \quad (o \le x \le b)$$

$$(13)$$

is solved for the Laplace transformations  $q_{m}^{g}$  (b,s) and  $q_{R}^{g}$  (0,s) of the time integrated transmission and reflection currents respectively. These last two quantities both have double poles at s = 0 (and also an infinite number of negative simple poles). These double poles at the origin lead asymptotically to a linear time behavior of  $Q_{m}^{g}(b,t)$  and  $Q_{m}^{g}(o,t)$ . By extrapolating these asymptotic solutions back to Q = 0, we obtain on the time axis as intercepts the group transmission delay times T<sub>R</sub> and the group reflection delay times T<sub>R</sub>. The method, though straight forward, leads to rather lengthy expressions for the higher group delay times [9], that will not be repeated here.

For the first group we list the following expressions (leaving out the group index 1 for simplicity):

$$q_{T}(b,s) = \frac{2\ell D\alpha}{s^{2}(\sinh \alpha b + 2D\alpha \cosh \alpha b)}$$
(14)  
$$q_{R}(o,s) = \frac{\ell(1-2D\alpha \coth \alpha b)}{s^{2}(1+2D\alpha \coth \alpha b)}$$
(15)

where  $\alpha(s) = (\{\Sigma + \frac{s}{v}\} / D)^{\prime 2}$  and  $\ell = \lim_{v \to 0} j^{+}(x,t) = \text{const. is}$  the fraction of the leakage current into the slab in group one once the omitted subscript is reinserted.

This leads to the asymptotic time behavior

$$Q_{TA}(b,t) = \lim_{s \to 0} \frac{d}{ds} (s^2 q_T(b,s)e^{st})$$
(16)

(a similar equation holds for  $Q_{RA}(o,t)$ ), which then gives

$$Q_{TA}(b,t) = j^{+}(b,\infty) (t - T_{T})$$
 (17)

$$Q_{RA}(o,t) = j^{-}(o,\infty) (t - T_R)$$
(18)

where

$$\mathbf{j}^{+}(\mathbf{b}, \mathbf{\infty}) = \frac{2\ell D}{L(\sinh \mathbf{b} + \frac{2D}{L}\cosh \mathbf{b})}$$
(19)

$$T_{T} = \frac{1}{2v\Sigma} \left\{ \frac{b}{L} \operatorname{coth} \frac{b}{L} \frac{(1+b+L}{(1+2D} \operatorname{coth} \frac{b}{L})}{L} - 1 \right\}$$
(20)

and

$$\mathbf{j}^{-}(\mathbf{o}, \mathbf{\infty}) = \frac{\ell(1 - \underline{L} \operatorname{coth} \underline{L})}{1 + 2\underline{D} \operatorname{coth} \underline{b}}$$
(21)

$$T_{R} = \frac{2T_{m} + (1 - \overline{L} \coth \overline{L})/v\Sigma}{1 - 2D \coth \overline{L}}$$
(22)

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Here 
$$L^{-1} = \lim_{s \to 0} \alpha(s) = (\Sigma/D)^2$$

Eqs. (19) to (22) are to be used for the first group in equations (9) and (10) to calculate the average delay times.

Note that for large coupling media, b>L, the transmission delay time in group one becomes a linear function of b, while the reflection delay time becomes a constant. Equations (20) and (22) show that  $T_T > T_B$ .

The complete 3 group formalism was applied to the coupling blankets of case I and case II and the results are included in TABLE I. Note that we have neglected the small fission rate in group one in the blanket in this treatment.

#### 3. SOLUTION OF KINETICS EQUATIONS

### 3.1. Analytical solution for step changes [9]

Calling the set of modules  $\{1, 2, \dots, j, M\} = S$ , an approximate analytical solution for step changes in the system (2) and (3) is found for  $t \gg \max(T_{jk}+T_{kj})$ . For smaller times, which are of no interest here, one would have to solve the system of differential-difference equations paying particular attention to conditions along the initial time lags [13]. Integrating the precursor equations directly to obtain

$$C_{ji} = \frac{\beta_{ji}}{\Lambda_j} \int_0^\infty T_j (t-T) e^{-\lambda} j i^T dT, \qquad (23)$$

letting all modules have equal amplitude time functions initially :  $T_1 = 1$ , the Laplace transformed system (3) after using (23) is written

$$H(s) T(s) = J(s) T_{a}$$
 (24)

where T(s) is the Laplace transform of the column matrix T(t) that has the elements  $T_j(t)$ . The matrices H(s) and J(s) are given by

$$H_{jj}(s) = s - \frac{\rho_{j} + \gamma_{j}}{\Lambda_{j}} + \sum_{i} \frac{s\beta_{ji}}{\Lambda_{j}(s + \lambda_{ji})} - \frac{\varepsilon_{jj}e^{-T_{jj}s}}{\Lambda_{j}}$$

$$H_{kj}(s) = -\frac{\varepsilon_{kj}e^{-T_{kj}s}}{\Lambda_{j}} (k \neq j)$$

$$J_{jj}(s) = 1 + \sum_{i} \frac{\beta_{ji}}{\Lambda_{j}(s + \lambda_{ji})} + \frac{\varepsilon_{jj}(1 - e^{-T}_{jj}s)}{\Lambda_{j}s}$$

$$J_{kj}(s) = \frac{\varepsilon_{kj}(1 - e^{-T}_{kj}s)}{s\Lambda_{j}} (k \neq j)$$

$$T_{i} = \frac{\varepsilon_{kj}(1 - e^{-T}_{kj}s)}{s\Lambda_{j}} (k \neq j)$$

If H(s) is non-singular then

$$T(s) = {H(s)}^{-1} J(s) T_{0}$$
 (25)

Let K(s) be the matrix of transposed co-factors of H(s); then (25) can be written

$$T(s) = \frac{K(s)}{|H(s)|} = \frac{W(s)}{|H(s)|}$$
(26)

where the column matrix  $\mathtt{W}(\mathtt{s})$  results from the indicated multiplications.

The inverse Laplace transformation of equation (26) gives the desired time functions

$$T(t) = \sum_{n} R_{n}(s_{n})$$
(27)

where the s are the roots of |H(s)| = 0 and where the  $R_n(s_n)$  are residues of  $T(s)e^{st}$  at  $s_n$ .

If  $W_1(s_n)$  and the derivative of  $|H(s_n)|$  with respect to s, denoted by  $|H(s_n)|'$  are both different from zero, then

 ${\rm T}(s)$  has a simple pole at  $s_n$  and the residue of  ${\rm T}(s)e^{st}$  at  $s_n$  is given by

$$R_{n}(s_{n}) = \frac{W_{j}(s_{n})e^{s_{n}t}}{|H(s_{n})|^{\prime}}$$
(28)

Note that W, and H(s) are analytic at s and that the singularities of T(s) and T(s)e<sup>st</sup> are the same since  $e^{st}$  is an entire function.

Thus, if the above conditions are satisfied, the amplitude function in each module is given by

$$T_{j}(t) = \sum_{n} A_{j}(s_{n})e^{s_{n}t}$$
<sup>(29)</sup>

where

$$A_{j}(s_{n}) = W_{j}(s_{n})/|H(s_{n})|^{\prime}$$
(30)

### 3.2. Influence of the delay times

Calculations for case I and case II of section 2.3 using the method described in the previous sections have been carried out.

If we assume temporarily that in these perturbations the coupling coefficients  $\varepsilon_{\rm m}$  and  $\varepsilon_{\rm R}$  are unchanged then we obtain the poles of the Laplace transformed solution, which are also the roots of the inhour equation of two coupled modules from |H(s)| = 0 in the form:

$$s(\Lambda + \sum_{i=1}^{\beta_{1}} (\rho_{1} + \gamma_{1} + \rho_{2} + \gamma_{2})/2 + (31) + \epsilon_{R} e^{-T_{R}s} \pm ([(\rho_{1} + \gamma_{1}) - (\rho_{2} + \gamma_{2})]^{2}/4 + \epsilon_{T}^{2} e^{-2T_{T}s})^{4}$$

There are 15 roots of eq. (31), that are qualitatively sketched by the solid lines in Fig. 2. (We get the same number of poles of the Laplace transformed solution if  $\varepsilon_{12} \neq \varepsilon_{21}$ .)

If one neglects time delays by setting  $T_{\rm n}$  and  $T_{\rm m}$  equal to zero in (31), then there are 14 roots R left as indicated by the broken lines in Fig.2. In particular, the largest negative root disappears and most of the other roots stay relatively unchanged. This indicates that the delay times will only have an influence on the solution in a very short time interval after the perturbation.

To quantify the argument we show in the first 3 columns of TABLE II the poles of the Laplace transform solution, which are the time eigenvalues of the system for case I unaltered, case I with delay times arbitrarily increased by a factor 10 and case I setting the delay times equal to zero respectively.



FIG.2. Roots of inhour equation for two cores (not to scale).

### TABLE II

Time	eigenvalues	case	Ι
------	-------------	------	---

asymptotic v	alues of coupl.	coefficients	critical values
regular	T'=TX10	T = 0	Τ = 0
-0.2687/8 <sup>a</sup>	-0.1545/7		
-0.2691/6	-0.1988/6	-0.281/6	-0.3104/6
-0.9418/4	-0.5367/4	-0.1027/5	-0.1779/5
-0.3201/1	-0.3201/1	<b>-</b> 0.3201/1	-0.3201/1
-0.3020/1	-0.3020/1	-0.3020/1	-0.3077/1
-0.1250/1	-0.1250/1	-0.1250/1 ′	-0.1249/1
-0.1046/1	-0.1046/1	-0.1046/1	-0.1106/1
-0.3225/0	-0.3225/0	-0.3225/0	-0.3223/0
<b>-</b> 0.1978/0	-0.1978/0	-0.1978/0	-0.2174/0
-0.1316/0	-0.1316/0	-0.1316/0	-0.1315/0
-0.5240/-1	-0.5240/-1	-0.5240/-1	-0.6609/-1
-0.3038/-1	-0.3038/-1	-0.3038/1	-0.3036/-1
-0.1355/-1	-0.1355/-1	<b>-</b> 0.1355/ <b>-</b> 1	-0.1386/-1
-0.1285/-1	-0.1285/-1	-0.1285/-1	-0.1285/-1
0.1854/0	0.1854/0	0.1854/0	0.4069/-1

a/-x means  $10^{-x}$ 

First note that the asymptotic system period, vis. the inverse of the positive time eigenvalue is unaltered within the accuracy of the calculation in the three cases. The main effect of increasing the delay times by a factor 10 is the reduction of the largest negative eigenvalue by about a factor 10. The poles contributed by the delayed neutrons are identical in the three cases. Case II and other examples have shown the same general behavior.

The plotted solutions for case I and case II are in Figures 3 and 4 in solid lines, where the options of using T = 0 and T' = 10T are indistinguishable from the plotted solution.







FIG.4. Amplitude functions: weakly coupled modules.

Thus we conclude that for excursions up to prompt criticality of the system, which are the most likely ones to occur in practice, the delay times may be neglected.

### 3.3. Influence of time dependent coupling coefficients

The influence of time dependent coupling coefficients will be inferred from a comparison of step input results using the asymptotic values for the coupling coefficients (see sect. 2.3.) referred to as time dependent coupling coefficients, to results using the critical values, referred to as time independent coupling. The input reactivity is the same in both calculations.

For strong coupling (case I) the time eigenvalues for asymptotic and critical values of the coupling coefficients are given in TABLE II, columns 3 and 4. Using the critical (time-independent) values, the asymptotic period of the system is underestimated by about a factor 5.

In Fig. 3 the amplitude functions for case I using the critical (time-independent) coupling coefficients are shown in broken lines. The underestimate of the period is quite obvious. There is also a 5% decrease in the amplitude function tilt.

For weak coupling (case II), which is not the case in proposed modular fast reactor cores, the influence of changing from time dependent to time independent coupling coefficients is, expectedly, not nearly as great, since the two modules behave more nearly like isolated reactors. Fig. 4 shows in broken lines the effect of time independent coupling again, which is still much more pronounced than any effects of the delay times.

In general the calculations have shown that the solutions for kinetics equations are quite sensitive to small changes in the coupling coefficients, and we conclude that in a more complete treatment, including feedback effects, the time dependence of the coupling coefficients must be considered.

### 3.4. Algorithm for numerical solution

Since the effect of the delay times has been shown to be negligible for times of interest, a numerical solution for the system (2) and (3) with all T=0 will be presented. It is based on an exponential approximation technique for point reactor kinetics by HANSEN [14]. A similar approach has been used by PLUTA et al [15].

For point reactor kinetics it is well known that conventional numerical techniques lead to exorbitant calculating times since the presence of a root at about  $-\beta/\Lambda$  in the inhour equation of the critical reactor requires very small time steps [16]. This problem is aggravated in coupled modular reactors and gets worse, the stronger the coupling, that is, the larger  $\varepsilon_{\rm T}$ . This can be seen by considering the largest negative root of the modular inhour

equation (eq.(31)) at criticality:  $\rho_1 = \rho_2 = 0$ ,  $\gamma_1 = \gamma_2 = \gamma_0$  and s  $\gg \lambda_i$ ; also remember that we consider T=0. This gives a root at about  $-(2\epsilon_m + \beta)/\Lambda$ . In our case I, which is representative of actual modular fast breeders,  $\epsilon_m = 5.7\beta$  (see section 2.3) and hence the most negative root is about a factor 10 larger in absolute value than for a single core fast reactor.

For the numerical solution of system (2) and (3) we define a column matrix  $\Phi$  with the elements  $T_1$ ,  $T_2$ ,  $C_{11}$ . ...,  $C_{16}$ ,  $C_{21}$ ...,  $C_{26}$ . We then write the system

$$\frac{d\Phi}{dt} = A\Phi$$
(32)
  
with
$$\frac{d_{1}}{d_{1}} = \frac{\epsilon_{21}}{\Lambda_{1}} + \frac{\lambda_{11}}{\Lambda_{1}} + \frac{\lambda_{16}}{\Lambda_{2}} + \frac{\lambda_{16}}{\Lambda_{21}} +$$

and 
$$d_j = \frac{\rho_j + \gamma_j - \beta_j + \varepsilon_{jj}}{\Lambda_j}$$

The matrix A is split into a diagonal matrix  $\underline{D}$ , a lower triangular matrix  $\underline{L}$  and an upper triangular matrix  $\underline{U}$ 

(33)

$$A = \underline{D} + \underline{L} + \underline{U}$$

The system (32) is then integrated over the time step h to yield  $\Phi(t+h) = e^{\underline{D}h} \Phi(t) + \int^{h} e^{\underline{D}(-h-\eta)} (L + \underline{U}) \Phi(t+\eta) d\eta$  (34)

To carry out the integration the column matrix  $\Phi$  in the integrand is approximated exponentially:

 $\Phi(t+n) = e^{\omega \eta} \Phi(t)$ (35)

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For step changes, the integration in eq. (34) is then carried out to yield:



where

$$g = \frac{\epsilon_{21}(e^{\omega_2 h} - e^{d_1 h})}{\Lambda_1(\omega_2 - d_1)}$$

and

$$g_{1j} = \frac{\lambda_{1j}(e^{\omega_{1j}n} - e^{\alpha_{1n}})}{\omega_{1j} - d_{1}}$$
$$g_{2j} = \frac{\lambda_{2j}(e^{\omega_{2j}n} - e^{\alpha_{2n}})}{\omega_{2j} - d_{2}}$$

The elements of the diagonal matrix  $\boldsymbol{\omega}$  are calculated initially from the previous time step

$$\omega_{j} = \frac{1}{h} \ln \left( \frac{\Phi_{j}(t)}{\Phi_{j}(t-h)} \right)$$
(37)

and are then iterated upon in the current time step until they satisfy a prescribed convergence criterion.

A numerical solution of this type was obtained for case II and is identical with the solution presented in figure 4.

For other than step change problems elements of matrices  $\underline{D}$ ,  $\underline{L}$  and  $\underline{U}$  become time dependent, and instead of eq. (34) we get:

 $\Phi(t+h) = e_{\theta}^{h} \underbrace{\mathcal{D}(\eta) \, d\eta}_{\Phi(t)} + \int_{0}^{h} e_{t+\eta}^{t+h} \left[ \underline{L}(t+\eta) + \underline{U}(t+\eta) \right] \Phi(t+\eta) \, d\eta$ (38)

#### 4. CONCLUSIONS

Based on analytical solutions of modular fast reactor kinetics equations valid for times larger than the delay times for neutron exchange between the modules, we conclude that for the class of transients below prompt critical the effect of the delay times is negligible. They thus do not have to be included into a model for the numerical integration of coupled module kinetics equations.

Solutions of the modular core kinetics equations are very sensitive to small changes in the coupling coefficients. This sensitivity increases with the size of the coupling coefficients and is quite pronounced for the strong coupling in modular fast breeder cores. Perturbations that produce neutron flux shape function tilts change the coupling coefficients. The time dependence of the coupling coefficients must thus be considered in the solution of modular core kinetics equations.

The computations have been carried out on the IBM 7094 of the Data Processing Center of Texas A&M University.

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### DISCUSSION

H.W. KUSTERS: In applying your method to coupled reactors you assume a step-wise perturbation in one or both of the modules, whereas some sort of ramp-rate reactivity insertion would be closer to reality. Can your method deal with such ramp-rate insertion and what effect would this have on the time dependence of the coupling coefficients, which in turn would considerably influence the excursion behaviour?

W.H. KÖHLER: The main part of our paper is a study of some of the problems involved in modular core kinetics. The ramp problem, or any other problem closer to reality, would have to be dealt with by the numerical solutions also outlined in the paper. One important point that we observed on looking at the simpler problems is the existence of a much more negative root than  $\beta/\lambda$ ; this leads to problems with the numerical solution, but we are better equipped to deal with these problems now. Since we have also noted the extreme sensitivity of coupling coefficients to changes in the flux shape, in the ramp problem we shall calculate coupling coefficients from static calculations before undertaking the main calculation and then check whether the evolving flux tilt will justify the use of these coefficients.

B.I. SPINRAD: Am I correct in assuming that the extra mode you get is due to the possibilities of symmetric and asymmetric perturbation?

It stands to reason, heuristically, that if one takes an initial critical system which is asymmetric, any perturbation can be taken as a linear superposition of the symmetric and an asymmetric perturbation.

W.H. KÖHLER: We did not use two modes simultaneously in this calculation; instead we calculated the coupling coefficients using the critical flux and the asymptotic lambda-mode flux in turn. In general, with coupled reactor kinetics, one exchanges difficulties in the calculation of higher mode contributions in a modal treatment for difficulties in calculating the time-dependent coupling coefficients. Perhaps a variational combination of two modes might be valuable in this respect.

### GENERAL DISCUSSION

F. STORRER (Chairman): I think it would be worthwhile to consider for a moment our motivation – as experimentalists and theoreticians – in doing these kinetic experiments on pulsation, modulation, correlation and what I shall label generally as reactor 'noise'.

Three motives come to mind. Firstly, I think we can readily agree that here theoreticians and experimentalists have come across an immensely important and fascinating field of study. Secondly, from these experiments and related theory we gain a better basic knowledge of the neutron processes occurring in a reactor. It is a fundamental tenet of pure research that it has no exact practical objective, but that it leads almost always to results of practical significance. I believe that in the case of critical experiments this general principle holds. We cannot overestimate the amount we have learned from studies of the techniques referred to in this session about neutron processes described by the Boltzmann equation. We are witnessing a dissection of this equation. together with the revelation of the spectrum of eigenvalues of the transport operator and associated eigenfunctions. There is no doubt that this information is of great practical use. Thirdly, one can think of the shortterm practical objectives - what results we can expect from these experiments that will be of direct use in our study of power reactors.

I would suggest that we start our discussion by considering what other objectives are being pursued in the various research centres, leaving aside the work presented at this Symposium. For instance, I wonder whether it is planned to make a systematic comparison of the calculated and measured values of the decay constant of the prompt neutrons in an assembly, and to introduce this comparison into the cross-section modification processes that have been described in Session III.

W.B. LOEWENSTEIN: The fast reactor community generally tends to disagree on the reliability of determinations of the effective delayed neutron fraction. I believe that many of the techniques described in these papers on kinetics might possibly provide a basis for an absolute determination of this parameter, because our current understanding of this is largely based on knowing the delayed neutron yields, as determined primarily from fission spectra, and then inserting them into a calculation. It is very difficult to compare theory and experiment at the present time without knowing the delayed neutron fraction reliably, so perhaps an effort to determine this in an integral fashion would be very productive.

G.S. BRUNSON (Scientific Secretary): The Agency held a small panel in the spring of 1967 on the subject of delayed neutrons, and one of the points it discussed then was the question of whether delayed neutron time curves exhibited growth-decay characteristics. There was one school of thought that felt that current delayed neutron data were really satisfactory, but on the other hand Dr. Tomlinson, from Harwell, stated that on the basis of separations he felt certain that about half of the 55-sec delayed neutron group did come through a growth-decay history; that is, half of it behaved in accordance with the Keepin data and another half of it grew in and then died out. This, therefore, raises the question as to whether the delayed neutron information, not only quantitatively but in time, is good enough for reactors with circulating fuel, or some sort of fast-start rocket reactor. D.STEGEMANN: This is more relevant to differential data on  $\beta$  than to time-dependent experiments. I agree that investigation of  $\beta_{eff}$  in integral assemblies, by a combination of either pulse source or noise measurements and static measurements of absolute reactor power, is of prime importance. We have carried out reactor measurements on STARK, comparing absolute reactor power measured by foil activation and fission rate traverses with measurements made by noise analysis, and these agreed fairly well, so we plan to continue this work now in fast systems although we cannot give any indication of the accuracy with which  $\beta_{eff}$  can be measured. I also believe that the application of kinetic measurements to noise analysis offers promising research prospects, and kinetic methods could be valuable too, as a diagnostic tool in the operation of power reactors.

B.I. SPINRAD: I should like to make two points. Firstly, there is always the problem of correlating the denotation of reactivity in terms of delayed neutrons (in dollars  $\beta_{eff}$  units) with its denotation in terms of absorption units (k units). In this regard, it is well to remember that the best approach is to make a good measurement of the flux and multiplication, which is a standard measurement and not so difficult as most kineticists seem to think. It is not valid to say that we do not know  $\beta_{eff}$  because we cannot measure it by strictly dynamic methods.

Secondly, I notice that most of the experimental work is done in terms of negative reactivities. It is well known, however, that the kinetics equations are not symmetric about criticality, so I think a good deal more investigation into both the experimental and theoretical measurement of kinetic parameters in rising period systems would be worthwhile.

A. FABRY: On the basis of recent exact integral measurements there is good reason to believe that the real fission spectrum of  $^{235}$ U is harder than that predicted by differential measurements, based usually on the proton recoil. This in turn influences the neutron lifetime in very hard fast reactors, as has already been shown. I should like to know what, if anything, should be done about this, and whether this type of error could occur in the fission spectra of more dilute fast reactors.

F. STORRER: In fact, we have been wondering whether the fission spectrum tail towards the lower energies was not actually greater than is generally accepted, and whether this modification of the fission spectrum would not reduce the discrepancy between calculated and measured neutron spectra.

R. VIDAL: I should like to add a little to Mr. Stegemann's comments to the effect that noise measurements on power reactors enable us to deduce the transfer function at different power levels. Measurements of this type have been done in France on Rapsodie, along with other critical measurements to obtain the transfer function by making sinusoidal reactivity perturbations at different frequencies and by analysing reactor response to reactivity jumps.

Y. YEIVIN: Reverting to the Chairman's introductory remarks, I wonder whether anyone would care to comment on the accuracy of kinetic measurements used as integral data in cross-section adjustment procedures, since this will emerge as the most important single factor in this context.

F. STORRER: We have been studying this at Cadarache, but it is still too soon to draw quantitative conclusions. Besides the spectral measurements, I think that we could usefully employ the experimental

### GENERAL DISCUSSION

value of the decay constant  $\alpha$ , for a fundamental or pseudofundamental mode, in a critical or sub-critical assembly. Preliminary calculations have shown that  $\alpha$  is often more sensitive than the reactivity or the material buckling to cross-section fluctuations at low energies. These results have still to be compared with the accuracy of experimental determination of these parameters. One should not, of course, anticipate the conclusions to be drawn from these studies, but it would be a shame not to draw on certain data already available to us.

On the other hand, while we believe we can make use of the  $\alpha$  values of the fundamental or pseudofundamental mode – which is an inherent characteristic of the system for a very large range of initial conditions – it seems that a detailed analysis of the transient behaviour would be difficult to carry out, since detailed calculation of the harmonics calls for an exact knowledge of the source (space, energy, time) and of the detector.

W.H. KÖHLER: My question is not related to kinetics. I should like to hear whether anyone here has done calculations on the photofission contribution in typical fast breeder reactors. I wonder whether this is, say, a  $\frac{1}{2}$ % contribution, or much less.

K.E.J. WIRTZ: I think much less; most of the capture gamma-rays will appear as cascades, and very few gamma-rays with energies in excess of 5 MeV will appear.

# SYMPOSIUM ON FAST REACTOR PHYSICS AND RELATED SAFETY PROBLEMS

## HELD AT KARLSRUHE, 30 OCTOBER - 3 NOVEMBER 1967

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