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FISSION PRODUCT NUCLEAR DATA (FPND)

VOL.II

PROCEEDINGS OF A PANEL
ON FISSION PRODUCT NUCLEAR DATA
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FOREWORD

Recent reactor safety investigations have accentuated the importance of fission products and intensified the need for detailed studies of their nuclear properties. This led the IAEA, under the guidance of its International Nuclear Data Committee, to convene a Panel Meeting of specialists to review, for the first time, the requirements for fission product nuclear data (FPND) in the light of present knowledge. The size of the meeting which was attended by more than 60 participants from 17 Member States and three international organizations illustrated the importance and magnitude of national efforts spent on FPND research. Sixteen internationally coordinated review papers distributed to participants before the meeting covered all aspects of use, status and requirements of FPND. They formed the basis for stimulating discussions between users, measurers and evaluators of FPND and led to numerous recommendations regarding future FPND work.

The proceedings of this panel are published in two parts. Part 1 consists of two volumes and contains all the review papers presented at the panel; the sequence of the paper numbers does not correspond to the sequence of their presentation (see panel programme). The historical development that led to the holding of this meeting and the scope of the panel are set out in detail in the "Introduction" (review paper no. 1a). A detailed summary of the panel's observations, conclusions and recommendations is given at the end of volume 2 and is followed by detailed tables in which required and achieved data accuracies are compared (Appendices A1-A5). Part 2 of the proceedings (volume 3) contains selected contributions to review papers. This part is being distributed only in a limited number of copies. The individual contributions are referred to at the end of review papers.

The scientific secretaries wish to express their deep appreciation to the panel participants for their very efficient cooperation during the meeting as well as thereafter during the preparation of the proceedings.

LIST OF ABBREVIATIONS

BWR	=	Boiling water reactor
CCDN	=	Centre de Compilation des Données Nucléaires
	=	NDCC = Neutron Data Compilation Center (of NEA)
EANDC	=	European-American Nuclear Data Committee
FP	=	Fission Product
FPND	=	Fission product nuclear data
FRO	=	Swedish zero power fast reactor
HTGR	=	High temperature gas cooled reactor
IAEA	=	International Atomic Energy Agency
INDC	=	International Nuclear Data Committee
LMFBR	=	Liquid Metal fast breeder reactor
ND	=	Nuclear data
NDS	=	Nuclear Data Section of the IAEA
NEA	=	Nuclear Energy Agency
ORELA	=	Oak Ridge linear accelerator
PFR	=	Prototype fast reactor
PWR	=	Pressurized water reactor
RP	=	Review paper
RPI	=	Rensselaer Polytechnic Institute
WRENDA	=	World Request List for Neutron Data Measurements

Panel on Fission Product Nuclear Data, Part I:

REVIEW PAPERS AND OBSERVATIONS, CONCLUSIONS AND RECOMMENDATIONS OF THE PANEL

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Note:
Volume 3 of these Panel proceedings contains "selected contributions to Review Papers", as indicated at the end of each review paper.

Review Paper no. 12

STATUS OF DECAY DATA OF FISSION PRODUCTS

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SUMMARY

The status of our present knowledge about fission product decay properties, i.e. half-life, mode of decay, beta- and gamma-ray energies and intensities, etc., is discussed. Compilations of such nuclear data are reviewed, and some recommendations are given.

Contributions from Aten and from Large have been incorporated into the review, and reference is made to a number of other contributions to the panel.

1. THE EXPERIMENTAL SITUATION

1.1. Introduction

Fission has now been known for about 35 years. During all these years an extensive work has been devoted to characterizing the fission products. To-day we can say that we know very much about the more long-lived species among those products. If a nuclide has a half-life in the order of hours or more, it is not difficult to design efficient chemical separation and purification procedures, to prepare a suitable radioactive source and to study the decay properties of the components in the source with various kinds of measuring techniques. These methods are therefore well established by now, and there is little point in discussing them here except, maybe, to point out that Ge(Li)-detectors have made most of the earlier work obsolete.

The situation is completely different when it comes to short-lived fission products. It is really amazing that so many fission products have been left unmeasured, or with very incompletely known properties, until now. It is true that the task of studying short-lived fission products is difficult, especially for half-lives in the second or subsecond region, but also many nuclides with half-life in the region of minutes are badly known, and part of the data available is seemingly erroneous.

One may, of course, ask the question whether short-lived fission products are of any interest whatsoever. Because of the short half-life their importance as tracers, for medical use, etc. is very limited. The application of our knowledge about them must be sought on the nuclear power side (for example: decay heating as a function of cooling time; effect in connection with possible reactor accidents). Quite generally, it seems fair to state that a strong motivation for a thorough study of the properties even of short-lived fission products is the fact that they are produced in large and increasing quantities as a result of the rapid development of nuclear power envisaged in many countries. It is also appropriate to point out that the scientific interest of these short-lived nuclides is very great. First of all, our knowledge about the structure of nuclei far away from the beta-stable region is quite rudimentary. Short-lived fission products are the best medium-mass probes on the neutron-rich side of stability, and it is therefore essential to study them thoroughly. There are also applications where their properties are extremely important, for instance in the astrophysical field. A proper treatment of the "r-process" in nucleosynthesis requires knowledge of nuclear half-lives, masses, beta-strengths, and the emission of delayed-neutrons. So far, one has to rely on extrapolations from what we know about nuclei close to the region of beta-stability (cf. mass formulas). Evidently, we can never do without extrapolations, because data are needed for nuclear species far more neutron-rich than the fission products but, using data for the most neutron-rich fission products which can be made, the extrapolations should become more accurate than they are at present. Thus, from the scientific point of view it is not hard to find reasons for studying extremely short-lived fission products.

1.2. New techniques

Until now we have had to rely mainly on clever chemists able to work out suitable separations methods for the fission product

elements. This problem has not been easy. The method has to be rapid and yet very selective with large decontamination factors from many elements. These are not the only difficulties, however. The sample prepared will, as a rule, still contain many isotopes of the desired element, and daughter activities will grow in. A gamma-spectrum of such a sample will therefore generally contain gammas from many components and, even if ingenious timing methods are used, the proper assignment of the gamma-rays to given nuclides is a difficult task. For these reasons rapid chemical separation methods and subsequent decay measurements are difficult to use for comprehensive nuclear studies, and only scattered data have come out for the more short-lived species.

It seems that the best way out of the experimental problems is to use some on-line system which continuously singles out the desired product and thus allows one to measure a steady-state sample at leisure just as one measures long-lived samples. The most perfect experimental set-up would then be one which separates out the desired nuclide in a monoisotopic form, i.e. accomplishes both element separation of mass separation. The sample should be free from disturbing contamination from all other fission products, and the separation should take place without any serious loss of activity and without any appreciable delay between production and sample preparation. The sample should, in addition, be obtained directly in the measuring position of the spectrometric apparates and in a form suitable for the measurements.

As usual, the perfect experimental set-up does not exist, but there are now several facilities in use which satisfy at least some of the requirements given above. This is not the place to describe in detail the various methods. As an illustration, however, the ISOL-facility at Studsvik will be briefly discussed. It consists of an isotope separator with its ion source close to the core of a 1 MW reactor in a position where the thermal neutron flux reaches 4×10^{11} n/cm^{2}sec. The target material, consisting of about 2 g of ^{235}U as oxide, is enclosed in the discharge chamber of the ion source. The ion source is kept so hot that many of the fission product elements diffuse through the target material and evaporate into the gas phase of the discharge chamber. Part of the evaporated atoms will get ionized and pass through the separator to the collector chamber where the ion beams are focussed. The radioactive atoms can then be collected directly in front of the measuring equipment.

This arrangement does not fill all requirements listed above. First of all, the delay, although short, is still too long for the study of many isomeric states. This is because the diffusion in the target material takes time. The most short-lived species studied so far is 0.12 s ^{132}In. For such a case one has to count on a severe decay loss but the high neutron flux and the large amount of target material ensures quite decent sample strengths (disintegration rates up to millicuries) for many short-lived activities. Another shortcoming is that the samples may contain several isobars owing to the fact that a large number of elements (all elements from zinc to strontium, except selenium,and those from silver to barium) are evaporated and ionized in the source. Often, this is not a serious problem because of the great difference in half-life from isobar to isobar and, besides, the mass separation can be combined with a rapid chemical separation step for identification purposes. In spite of these drawbacks, the facility is a convenient tool for the study of a large number of short-lived fission products, and an extensive programme for investigating their nuclear properties is being carried out.

This description of a particular arrangement for the study of short-lived fission products is only meant to exemplify the techniques now available. Other facilities have other advantages and disadvantages. An intensive experimental work on these nuclides is being pursued in various places, however, and it is to be expected that our knowledge about the properties of the short-lived fission products will rapidly improve in the next few years to the benefit both of basic science and of the applied field.

It should be pointed out that rapid chemical separation methods still have their place in the study of short-lived nuclides for providing complementary information.

1.3. New experimental results

Using the new techniques the collection of new fission product nuclear data is in rapid progress in various laboratories. Many results have been published already and are thus readily available. They have also been included in the most recent compilations. There is also a wealth of data lying around unpublished and therefore not so readily available, however. Many scientists in the field are so occupied doing

experiments that they hardly give themselves time to write up the results. They may also by trying to add just one more piece of evidence for a particular level scheme when the results already obtained are perfectly valid for the applications which we discuss here.

Thus, it might be difficult to squeeze the data out of the scientists at an early time. There lies also a danger in trying to compile as fresh data as possible, because the results are then often in a preliminary form and not analyzed well enough to warrant a wide spreading. Erroneous results might creep into the compilations, and it might be hard to get them out again. Thus, the compilers and evaluators must be cautious when compiling results in a premature form. For the same reason, data presented in informal conferences and perhaps reproduced in un-edited proceedings must also be treated with care.

2. ACCURACY OF FISSION PRODUCT NUCLEAR DATA

2.1. Half-life determinations

It is not easy to correctly assign errors to measured half-life data. The reason for this is not the statistical treatment of the measurements, which anybody can do without difficulty, but rather various kinds of systematic errors. Half-life determinations of long-lived nuclides might suffer from long-time instability of the measuring equipment, not properly taken care of by monitoring procedures, or from some minute contamination which is very difficult to detect. One should preferably follow the decay for many half-lives checking that the measured points are properly distributed in order to make sure that there is no contamination in the sample, and this is not always practical.

Similar difficulties appear when measuring short-lived samples. Problems with the long-time stability are not severe here, but the contamination problem might be worse. Usually,there are several components in the sample, and the composite decay curve must be resolved. One must not get fooled by the fact that the mathematical treatment in such a case often gives an excellent result with a very small statistical error. The inclusion of still another component of intermediate half-life into the analysis can drastically change the results. Therefore, the analysis must be checked very carefully searching systematic trends in the data which might reveal contamination.

Because of unknown systematic errors one should be very suspicious when half-lives are quoted with errors less than about 1 %. This is not to say that half-life determinations cannot be made better than that, but such determinations require very careful work.

Very often several half-life determinations are presented by the same author. The result might look very accurate but one must then remember that the same kind of systematic error is likely to appear in all the measurements. A better test is to compare determinations made by different groups using different sources and techniques. In doing so, one often finds that the results do not overlap very well, obviously because of systematic errors not taken into account.

What has been said above goes mainly for beta measurements. If the half-life is determined in experiments where the decay of a certain gamma-ray is measured with Ge(Li)-detectors the problem with contamination will be smaller. Such measurements often suffer from low counting rates, however, and sometimes it is not easy to properly subtract the background of Compton events from other gamma-rays in the sample. Also, the dead time of the system must be accurately determined.

2.2. Gamma-rays

With good Ge(Li)-spectrometers at hand the accuracy in gamma-energy determinations is often excellent. This is fortunate because one can often use a well-determined gamma-ray as a label for a given nuclide The intensity is more difficult to measure accurately, however. It requires a well-calibrated spectrometer with due account taken of escape probability, summing probability, etc. In a contribution to this paper Large points out some problems encountered in the evaluation of gamma-ray energies and intensities. This part of his contribution is here reproduced in extenso:

(N.R. Large, Harwell)

"In any evaluation of γ-ray energies and intensities difficulties are encountered in attempting to establish reliable values for the absolute intensities. There is usually little difficulty in establishing the energies with fairly high precision, and the relative intensities are often known with reasonable accuracy. Conversion of relative intensities to absolute intensities can only be carried out if the β-branching ratios are known; in particular the intensity of the β-branch to the ground state

is required. Unfortunately, the intensity of this β-branch is usually the most difficult to determine. In many cases the β-branching ratios to the remaining energy levels are calculated from the measured relative γ-ray intensities on the basis of a postulated decay scheme, but no information on the ground state β-branch can be obtained in this way unless other experimental evidence on β-branching is available. The resulting uncertainty in the intensity of the ground state β-branch is in many cases the major contributor towards the uncertainty in γ-ray intensities.

Another source of difficulty in evaluating information on γ-ray intensities lies in the uncertainty, and in some cases confusion, with regard to conversion coefficients. Here there are several problems. Firstly, a few authors fail to make clear in table headings the distinction between γ-ray intensities and γ-transition intensities, and others do not make clear whether their conversion coefficients are for the K shell only or for all electron shells. In these cases it is necessary to study the papers in great detail and often to find alternative sources of information in order to clear up the ambiguity. The second problem arises from the fact that in a majority of cases only K-conversion coefficients are measured, and values for L- and M-conversion coefficients have to be obtained by interpolation in tables such as that by Hager and Seltzer in Nuclear Data Tables. For the purpose of this interpolation assumptions must be made about the γ-transition type unless this has been determined by the authors. Where this procedure has been carried out by the authors the evaluator is relieved of the task, but uncertainties are still introduced into the absolute intensity data if highly converted transitions have to be taken into account in order to obtain an intensity balance. There are, of course, cases where no attempt has been made to determine any conversion coefficients, and in these cases the only way of dealing with the problem is to make some reasonable assumptions about transition types and to obtain all conversion data from tables - this can lead to large errors.

It will often be found that where conversion coefficients are quoted these have not been measured, but depend on separate relative intensity measurements on γ-rays and conversion electrons, coupled with a theoretical value for the conversion coefficient for one transition for which a transition type is assumed. This value enables the remaining intensities to be normalised, and the consistency of the overall decay scheme is taken as support for the original assumption."

An effort to measure absolute intensities of gamma-rays with high accuracy is described by Debertin et al in a contribution to this review paper.

2.3. Beta-rays and Q-values

A completely known decay also implies that the beta branch feeding each level in the daughter nucleus is known. A direct measurement of the beta branches is a tedious and difficult task, however, especially for short-lived nuclei with high Q-values and many beta branches. It can be done by measuring, in coincidence arrangements, the beta-branches feeding the different energy levels. In addition, the ground state branch must be measured. Usually, however, the beta branching ratios are inferred indirectly from the level scheme and the measured gamma transition rates. From the errors in the gamma transition rates the error of the beta branch can be calculated, but it is difficult to judge such errors. The beta branches and their errors depend completely on the level scheme chosen. If this should be wrong, the beta intensities are also wrong. Thus the errors assigned are, in a way, meaningless unless the level scheme is definitely proven to be correct - a formidable task for more complicated cases.

There is an alternative way to study the beta intensities without knowing the decay scheme in detail. It is a crude method which only can reproduce the gross pattern of the beta branch intensities, but it can be used also for very short-lived nuclei with high Q-values. I refer to the measurement of the beta strength by means of a "total absorption"-gamma detector, i.e. large gamma-detectors, usually in a 4 π-geometry, with large probability of summing the various components in a gamma cascade. Results for 34 neutron-rich nuclei with half-life down to about 30 sec have been published [1], and a new measurement comprising close to 50 new cases (half-life down to about 1 sec) is under way [2]. The beta strength function is determined for each particular nuclide. It can easily be transformed into the beta feed function (i.e. probability per energy unit of beta decay as a function for the excitation energy of the daughter nucleus), which is the important function for various applications.

The error to be assigned to the beta strength is difficult to estimate. It would be simple with a perfect total-absorption spectrometer. The ones used are far from perfect, however, and the accuracy then de-

pends on converting the pulse spectrum to beta feed spectrum. This, in turn, depends on theoretical assumptions concerning the development of the gamma-cascades. Thus, an appropriate experimental error cannot be given.

Mere Q_β-values are perhaps of minor interest for the various applications of fission product data under discussion here. On the other hand, knowing the Q_β-values in a systematic way one can construct the nuclear mass surface, and this is of paramount importance for many scientific applications. It is therefore not out of place to discuss them.

Most Q_β-values close to the beta-stable region are well known and need no discussion. For short-lived fission products the situation is quite different. Until recently they have been almost unknown, but systematic studies are now under way [3,4]. The technique employed is to measure the beta spectrum coincident with a gamma gate corresponding to an excited level. The sum of the beta end-point energy and the gate energy will then be a measure of the Q-value if it can be proven that the beta branch measured really feeds the level used as gate directly (otherwise the sum will be a lower limit of the Q-value), and the error of the measurement is composed of the errors of the beta and gamma measurements. The accuracy varies from case to case and is usually in the range 20 - 200 keV.

3. REVIEW OF AVAILABLE DECAY DATA COMPILATIONS

3.1. Introduction

After this general discussion about the experimental techniques and results some compilations containing fission product nuclear data will now be reviewed. Because of the rapid development of the field in the last few years most of the older compilations are obsolete or, at any rate, very incomplete. For this reason compilations older than 3 - 4 years have been left out.[x] Nor will nuclear charts be discussed. They are very handy for a quick look at the decay properties, especially the half-lives, but they cannot possibly contain enough data to satisfy one's needs.

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x) Reference is made to the "List of Compilations, Evaluations, and Computer Codes of F.P.N.D." prepared for the panel, for compilations not treated in the present review.

A special place among the compilations is taken by the periodicals Nuclear Data Tables and Nuclear Data Sheets. They are both very important. The Nuclear Data Sheets contain comprehensive and evaluated compilations of nuclear data for isobars of certain mass numbers. The advantage of this procedure is that a self-consistent picture is produced with all the available detail. The disadvantage is that the period which elapses between successive evaluations for a given nuclide is such that for many nuclides the evaluations are very considerably out of date, and the data are therefore only of limited value to users who need to cover the whole range of fission products. In order to remedy this, reference lists are periodically issued so that one can update the information for a particular mass number starting from the latest compilation for this mass number.

Generally, the compilations try to cover the whole range of fission products with emphasis on certain decay properties. A thorough check of the uptodateness and the completeness of the various compilations is too time-consuming to be carried out here. What one can do within a reasonable amount of time is, however, to check whether a few articles, chosen more or less at random, have been taken into account. Such checks were carried out using as test cases the half-life of ^{132}Sn for which the experimental information is contradictory with one value of 2.1 ± 0.2 m given by Lin and Wahl in 1970 [5] which is in accord with an old determination of 2.1 m from 1956 [6], and a number of determinations pointing at a half-life of 40 s (Strom et al (1966): 60 ± 10 s [7], Nunnelley et al (1972): 40.6 ± 0.8 s [8]; Neumann et al (1972): 39.0 ± 1.0 s [9]; Kerek et al (1972): 40 ± 1 s [10]). Another test case is the article by Carlson et al [11] containing many well-determined half-lives for short-lived xenon and cesium isotopes, e.g. ^{140}Xe: 13.60 ± 0.10 s; and ^{140}Cs: 63.7 ± 0.3 s. Test cases for gamma-rays have been ^{80}As (McMillan and Pate (1971) [12]) and ^{125}Sn (Wild and Walters (1967) [13]; Macias and Walters (1971) [14]).

3.2. R.H. Filby, A.I. Davis, K.R. Shah, G.G. Wainscott, W.A. Haller, and W.A. Cassatt, Gamma Ray Energy Tables for Neutron Activation Analysis, WSUNRC-97(2) (1970)

Gamma-ray energies are ordered by increasing energy both in a general list and in lists for each nuclide. Limits of error are not given; nor are the measurements from several sources averaged. The literature

is said to have been followed until 1969, but for the test case ^{125}Sn the 1967-year reference |13| was not used but apparently an older reference from 1964 (quoted in Tables of Isotopes 1967 |15|).
Although one should not put too much weight on a single case, there are apparently some doubts about the completeness of the compilation.

As the compilation is intended for neutron activation analysis work, only certain fission products are included. This limits its general usefulness.

3.3. Chr. Meixner, Gammaenergien Teil I (Jül-811-RX), Teil II (Jül-812-RX) Teil III (Jül-813-RX), (1971)

"Teil I" contains gamma energies ordered after atomic number of the decaying nuclide. The energies have been evaluated, and average values with their errors have been calculated.

The compiler has been mainly interested in activation analysis, and he states that the emphasis of the work is directed towards products from neutron-induced reactions. Nevertheless, the choice of nuclides treated seems to be rather arbitrary. ^{125g}Sn is not found in the compilation, nor is ^{80}As. On the whole, the compilation is quite incomplete.

Relative or absolute intensities (without errors) are given.

The table also contains schematic gamma spectra for the cases considered (i.e. a gamma-ray is shown as a line of length corresponding to its intensity).

"Teil II" and "Teil III" contain gamma-rays ordered after energy, the former for products from neutron-induced reactions and the latter for fission products. "Teil III" also contains nuclides ordered after half-life (without quotation of the error). The same data are used as in "Teil I", which means that the data are severely incomplete. Among the test cases only the half-lives of ^{140}Xe and ^{140}Cs are compiled.

The lack of completeness of this compilation is a serious drawback for those who are interested in fission products.

3.4. J. Blachot et R. de Tourreil, Bibliotheque de données nucléaires rélatives aux produits de fission, CEA-N-1526, 1972

This work contains information about half-lives, gamma-rays, total decay energies, delayed neutrons, etc. Limits of errors are not given, nor is the choice of particular data justified. The literature has been followed until the end of 1970.

All the test cases are included in the compilation.

The compilation is stored on computer-compatible magnetic tape, and very clear an easily readable print-outs can be distributed.

As for completeness this work seems to be excellent. Because of its computer-compatible form it would be easy to update it more or less continuously. The only drawback is that limits of error are lacking.

At the end of the compilation the gamma-rays are ordered according to increasing energy.

The compilation is described in a contribution to the IAEA-meeting in Paris, March 1973 [16].

3.5. A. Tobias, Data for the Calculation of Gamma Radiation Spectra and Beta Heating from Fission Products (Revision 3), CEGB/RD/B/M2669, 1973

This is very complete compilation, with all test cases included. It is a pity that limits of error are not given in this compilation, and that data from different experiments have not been averaged. The compilation is very recent covering the literature until March 1973. The fact that it is already the third revision of the original version, which appeared in 1970 [17], shows the intention of keeping the compilation very up-to-date. In addition to tables of gamma-ray energies and half-lives there is a table with estimated mean beta energies for decay heating calculations.

For a survey on the current status of fission product data, containing information about the compilation, reference is made to a contribution by Davies and Tobias [18].

3.6. M.J. Martin and P.B. Blichert-Toft, Radioative Atoms, Auger-Electron, α-, β-, γ-, and X-Ray Data, Nuclear Data Tables A8 (1970) 1

This compilation is restricted to radioactive atoms of special importance in nuclear medicine, in health physics, and for industrial applications. This means that it is very incomplete as regards fission products. For the nuclides considered, however, the treatment seems quite comprehensive, including level schemes, conversion electron data, etc., with limits of error given. The data are evaluated and, in case of several determinations, recommended values are chosen. For half-lives, for example, a reasonably complete literature survey has been made. The results of various determinations are shown, and weighted averages are calculated.

The compilation seems to be very thoroughly worked-out. One drawback is its incompleteness. Besides, it is rather old (literature covered until September, 1969).

3.7. M.A. Wakat, Catalogue of Gamma-Rays Emitted by Radionuclides, Nuclear Data Tables A8 (1970) 445

The gamma-rays are ordered after energy. Two associated gamma-rays are given for each case. The literature is surveyed until 1970. The limits of error are not explicitly given for each case but can be inferred from the number of significant figures. The test case (^{125}Sn) is included.

This is probably a fairly complete compilation.

3.8 O.J. Eder and M. Lammer, The Influence of Uncertainties in Fission-Product Nuclear Data on the Interpretation of γ-Spectrometric Measurements on Burnt Fuel Elements, IAEA/SM-170/12 (1973)

The paper contains, among other things, a description of a fission product nuclear data file at Seibersdorf. The file is restricted to long-lived nuclides (half-life in general longer than one day) which means that it is quite incomplete. For the fission products chosen, however, the treatment seems to be extensive with data from various sources compared and properly evaluated. The compilation contains gamma-ray energies and intensities as well as half-lives, with limits of errors given, for the various nuclides. A table of gamma-rays arranged according to energy is also given.

In a contribution to this review Lammer describes in more detail the evaluation of half lives and branching ratios and quotes the sources of data. The literature search was continous up to the time of the Panel. The gamma ray table is not published, but the treatment and sources of data are included in the contribution. A thorough literature search was done until the beginning of 1971, with frequent updates later on.

3.9. N.R. Large and R.J. Bullock, A Table of Radioactive Nuclides Arranged in Ascending Order of Half-Life, Nuclear Data Tables A7 (1970) 477

This compilation contains only the half-lives. No limits of error are given. As regards the test cases, the half-life of ^{132}Sn is given as 2.1 m but the value 60 s, published in 1966, does not appear. This arises some doubts as to the completeness of the compilation.

It is hard to see any real use of a compilation like this one. If one wants to make an identification of an activity only based on the half-life, the half-life determination will have to be very accurate. However, a comparison with the entries in the table is not very conclusive, as the limits of error are not stated there. A table of half-lives only should be based on an evaluation of published experimental values, and the appropriate accuracy should be stated. Otherwise, the value of the compilation is very limited.

3.10. L. Tomlinson, Delayed Neutrons from Fission. A Compilation and Evaluation of Experimental Data, AERE-R 6993 (1972)

This compilation belongs more to the paper about delayed neutrons. It will be briefly mentioned here also, however, because of its half-life evaluations, where the experimental data, with their errors, are clearly stated, and the mean values are calculated. The only remark to make is that the error of the recommended value is in a few cases possibly too small (Cf. Section 2.1). Is the half-life of ^{87}Br really known with an accuracy (standard deviation) of 0.2 %, for example?

The compilation is now being revised.

3.11. W.T. Bass, D.J. Horen, and W.B. Ewbank, Current Nuclear Level Schemes; A = 91 - 117, ORNL-4627 (1970)

D.J. Horen (editor), Current Nuclear Level Schemes: A = 118 - -139, ORNL-4730 (1971)

These compilations contain level schemes (as ladder diagrams) and tables of level energies and spins. Nuclear decay data such as half-lives, beta- and gamma-energies are not given. The level schemes are deduced from radioactive decay data but also from other kinds of experiments (inelastic scattering and Coulomb excitation, neutron capture, stripping and pick-up reactions).

The diagrams in the compilations are very illustrative and give the reader a quick and, as it seems, rather complete insight about what is known about nuclear levels. It is to be hoped that this kind of compilation is kept up-to-date by frequent revisions.

3.12 A.H. Wapstra and N.B. Gove, The 1971 Atomic Mass Evaluation, Nuclear Data Tables A9 (1971) 267

This compilation contains Q_β -values extracted from evaluations of available experimental data. The results are mainly based on measured data, but for some cases systematical extrapolations and interpolations are used. The compilation can be considered as a standard for the decay energies.

3.13 J. Mantel, The Beta Ray Spectrum and the Average Beta Energy of Several Isotopes of Interest in Medicine and Biology, Int. J. Appl. Radiat. Isot. 23 (1972) 407

From known beta-decay properties (branching ratios, end-point energies, forbiddenness) the composite beta spectrum is calculated using the Fermi theory, and the average beta energy is evaluated. The resulting spectra are given in graphs for some 60 nuclides, among them a few fission products. Thus, for fission products the compilation is quite incomplete, but it contains data which are very difficult to extract directly from experimental articles. Possibly, it would be worth while to extend this work to include more fission products.

3.14 Miscellaneous compilations

a) T.R. England, An Investigation of Fission Product Behavior and Decay Heating in Nuclear Reactors, thesis, University of Wisconsin Microfilm 70-12, 727 (1970):

This compilation can be used as a list of the beta- and gamma-energy release in the decay of various nuclides. Surprisingly enough the basic energy data and the half-lives used in the tables are very old (the newest ones from 1966), which limits the value of the compilation.

b) M. Sakai, Quasi,Ground, Quasi-Beta, and Quasi-Gamma Bands - 1972, Nuclear Data Tables A10 (1972) 511:

A list of probable members of quasi-ground, quasi-beta, and quasi-gamma bands of nuclei is presented. The compilation is of interest in the basic scientific field rather than in the applied field.

c) A.Z. Nagy, A. Csöke, and E. Szabo, Two-dimensional (T,E_γ)-Mapping of Short-Lived Nuclides, J. Radioanal. Chem. 7 (1971) 365:

A table is given containing half-lives and major gamma-energies (all without errors). The literature scanned is too old, however, the newest source being the 1967 Table of Isotopes. As a compilation the article is therefore obsolete.

d) M.E. Meek and R.S. Gilbert, Summary of Gamma and Beta Energy and Intensity Data, NEDO-12037 (1970):

The compilation concentrates on volatile fission products, i.e. essentially mass chains containing halogens and noble gases. For this reason it is quite incomplete. Most data are taken from reference [15] and thus quite old by now.

No limits of error are given for any quantity.

4. RECOMMENDATIONS

4.1. Choice between compilations

The compilation to choose depends, of course, on one's needs since the different compilations may concentrate on different aspects of the fission product decay. For general information, however, there are two compilations to be recommended, e.g. the one by Blachot and de Tourreil (3.4) and the one by Tobias (3.5). Both seem to be very complete as far as fission product nuclear data are concerned, and both are very recent. Also, one has reasons to believe that they will be kept up-to-date by frequent revisions.

Among more specialized compilations must be mentioned the 1971 Atomic Mass Evaluation (3.12) which is of great value for Q_β-values, and the very interesting approach by Horen et al: Current Nuclear Level Schemes (3.11).

In addition to the above-mentioned compilations, one should not forget consulting the Nuclear Data Sheets (3.1).

4.2. On the Evaluation of Experimental Results

One impression when looking through the various compilations is that evaluations are seldom presented in a clear way. Often only one figure is given for a certain quantity so that the compiler has obviously made his choice among the experimental results avialable, but he does not justify this choice. Morover, with a few exceptions, the limits of error are not given which is a great drawback for the reader who might wish to know what confidence to put into the given number without going to the original reference. Therefore, one would like to urge the compilers to improve this part of the presentation of their efforts. For many readers this would increase the value of the compilations considerably.

4.3. New Experimental Work Needed

In their analysis (reference [18]) Davies and Tobias arrive at the conclusion that our of 449 radioactive products there are 42 nuclides with one parameter poorly known, 110 nuclides with two parameters poorly known and 14 with three parameters poorly known (parameters: half-life; gamma-radiation; beta-radiation). In addition to this, there are many fission products which have not been studied at all. Thus, a great amount of experimental work is still needed before one can say that the decay pro-

perties of the fission products are known to a satisfying degree. The new techniques briefly touched in Section 1 of this paper will undoubtly fill many of the gaps, but for certain elements the experimental difficulties are very great. Therefore, one has to foresee a long time of tedious experimental work before a satisfactory situation is reached.

4.4. Errors in Published Fission Product Nuclear Data

Flynn, and others, have pointed out that published decay properties are sometimes obviously erroneous. It would be of great value to many users of fission product nuclear data to have access to lists of such cases, if possible also containing rectified values. It might well be the duty of the panel to collect information about fission products for which there are serious doubts concerning the correctness of the published data, and to relay this information to the users in a suitable way.

5. NEW INFORMATION

In a contribution to this paper Aten has pointed out the need for accurate information about decay properties of certain fission products of special importance for calibration purposes. Appendix A reproduces a table of suitable reference sources.

A survey of the short-lived activities obtained using the OSIRIS on-line isotope separator has been prepared [19]. With the results from this survey included an evaluation of half-life data for short-lived fission products in the mass ranges 74 - 80, 85 - 95, and 111 - 145 has been carried out. The results are given in Appendix B.

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Finally, three compilations containing decay data for fission products are reported to be under way although not yet available, namely:

R.L. Heath, AEC Catalog of Gamma Rays of Fission Product Data,

V. Sangiust et al, Gamma Spectra for Short-Lived Fission Products

and D.G. Vallis, Computerised Storage and Interpretation of Nuclear Decay Schemes.

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* See Appendix A.

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APPENDIX A (Contribution to Paper No 12 of the IAEA Panel on Fission Product Nuclear Data)

FISSION PRODUCTS WITH YIELDS SUITABLE FOR REFERENCE DATA

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Nuclide	t 1/2	Gamma-ray	Standard	Tracer
$^{91}Sr(+^{91}Sr^{m})$	9.7 hours	0.748 MeV 1.025	^{95}Nb 0.764 MeV ^{137}Cs 0.662 ^{65}Zn 1.115	^{85}Sr
$^{95}Zr(+^{95}Nb)$	65 days	0.724 0.756	^{95}Nb 0.764 ^{137}Cs 0.662	^{88}Zr
$^{97}Zr+^{97}Nb^{m}$	17 hours	0.747($^{97}Nb^{m}$)	^{95}Nb 0.764	^{88}Zr
$^{99}Mo+^{99}Tc^{m}$		0.140($^{99}Tc^{m}$)	^{139}Ca 0.166 ^{141}Ce 0.145 ^{57}Co {0.122 {0.136	
$^{103}Ru(+^{103}Rh^{m})$	40 days	0.497	^{85}Sr 0.514	^{106}Ru
$^{105}Ru(+^{105}Rh)$	4.4 hours	0.726	^{95}Nb 0.764 ^{137}Cs 0.662	^{106}Ru
^{105}Rh	40 hours	0.306 0.319		^{102}Rh
$^{106}Ru+^{106}Rh$	1 year	0.512(^{106}Rh)	^{85}Sr 0.514	^{105}Ru
$^{131}I(+^{131}Xe^{m})$	8 days	0.364	$^{113}Sn+^{113}In$ 0.393	^{125}I
		0.637	^{137}Cs 0.662	^{126}I
		0.723	^{95}Nb 0.764	
$^{132}Te+^{132}I$	78 hours	0.230 0.668(^{132}I)	^{137}Cs 0.662	
^{133}I	21 hours	0.53	^{85}Sr 0.514	^{125}I ^{126}I
^{134}Cs	2 years	0.605 0.796	^{134}Cs ^{95}Nb 0.764	^{132}Cs
$^{137}Cs+^{137}Ba^{m}$	30 years	0.662 ($^{137}Ba^{m}$)	^{137}Cs 0.662	^{132}Cs
$^{140}Ba+^{140}La$	13 years	0.537 1.598(^{140}La)	^{85}Sr 0.514 ^{40}K 1.460	^{133}Ba
^{141}Ce	33 days	0.145	^{141}Ce 0.145 ^{139}Ce 0.166	^{139}Ce
^{143}Ce	33 hours	0.293 0.668 0.725	^{137}Cs 0.662 ^{95}Nb 0.764	^{139}Ce
^{147}Nd	11 days	0.533	^{85}Sr 0.514	

APPENDIX B

EVALUATION OF HALF-LIFE DATA FOR SHORT-LIVED FISSION PRODUCTS

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A comparison between fission product half-life determinations obtained using the OSIRIS isotope-separator-on-line facility at Studsvik (References [2],[3],[28],[31],[34],[44],[45],[48],[51],[56]) and values published in the literature is presented below (the literature search might be incomplete, and it is left to the compilers in the field to make a more extensive evaluation). The mass ranges covered are 74 - 80, 85 - 95, and 111 - 145. The errors given are to be considered as corresponding to one standard deviation.

In the evaluation no measurement has been given a greater weight than that corresponding to a standard deviation of 0.5 % of the measured value. Also, the minimum "recommended error" corresponds to 0.5 % of the mean.

Mass number	Element	Half-life Measured	Reference	Half-life Recommended	Comment
74	Zn	98 ± 2 s	1	96 ± 1 s	
		95 ± 1 s	2		
75	Zn	9 ± 2 s	3	10.2 ± 0.3	
		10.2 ± 0.3 s	2		
75	Ga	120 ± 12 s	4	126 ± 2 s	
		126 ± 2 s	2		
76	Zn	6 ± 1 s	3	5.7 ± 0.3 s	
		5.7 ± 0.3 s	2		
76	Ga	27.1 ± 0.2 s	5	27.6 ± 1.1 s	
		29.8 ± 0.4 s	2		
77	Zn	1.4 ± 0.3 s	3	1.4 ± 0.3 s	
77	Ga	17.1 ± 1.5 s	6	13.2 ± 0.8 s	
		13.0 ± 0.3 s	2		

Mass number	Element	Half-life Measured	Reference	Half-life Recommended	Comment
77	Ge	52 ± 2 s	7	53.5 ± 0.6 s	
		53.6 ± 0.9 s	8		
		55.5 ± 1.0 s	9		
		52.9 ± 0.6 s	2		
78	Ga	4.8 ± 1.3 s	10	5.09 ± 0.05 s	
		5.09 ± 0.05 s	2		
79	Ga	3.00 ± 0.08 s	2	3.00 ± 0.08 s	
79	Ge	40 ± 4 s	11	40 ± 3 s	There are possibly two isomers of ^{79}Ge with half-lives 40 s and 19 s, respectively.
		41.1 ± 4.3 s	10		
79	Ge(?)	19.1 ± 0.3 s	2	19.1 ± 0.3 s	
80	Ga	1.7 ± 0.2 s	3	1.7 ± 0.2 s	
80	Ge	24 ± 1 s	3	26 ± 2 s	The discrepancy between different determinations is large. No weight factors are used in calculating the average value
		24.5 ± 1.0 s	10		
		29.5 ± 0.4 s	2		
85	Br	180 ± 3 s	12	175 ± 4 s	
		172 ± 2 s	2		
86	Br	54 ± 2 s	13	55.7 ± 0.5 s	
		59 ± 4 s	14		
		55.7 ± 0.5 s	2		
87	Br	56.1 ± 0.7 s	12	55.8 ± 0.3 s	
		55.8 ± 0.3 s	15		
		55.6 ± 0.2 s	16		
		56.3 ± 0.5 s	2		
88	Br	15.5 ± 0.3 s	12	16.0 ± 0.2 s	
		16.3 ± 0.8 s	17		
		15.9 ± 0.1 s	15		
		16.5 ± 0.2 s	2		
89	Br	4.4 ± 0.5 s	17	4.54 ± 0.10 s	
		4.5 ± 0.4 s	15		
		4.55 ± 0.10 s	2		
90	Kr	32.32 ± 0.09 s	18	32.2 ± 0.2 s	
		34 ± 3 s	2		

Mass number	Element	Half-life Measured	Reference	Recommended	Comment
91	Kr	8.57 ± 0.04 s	18	8.7 ± 0.2 s	
		9.16 ± 0.09 s	2		
91	Rb	58.2 ± 0.2 s	18	58.2 ± 0.3 s	
		58.2 ± 1.0 s	2		
92	Rb	4.50 ± 0.03 s	18	4.50 ± 0.03 s	
		4.50 ± 0.04 s	2		
93	Rb	5.86 ± 0.13 s	18	5.81 ± 0.04 s	
		5.8 ± 0.1 s	19		
		5.80 ± 0.05 s	2		
93	Sr	7.32 ± 0.10 m	18	7.36 ± 0.15 m	
		7.43 ± 0.03 m	20		
		8.22 ± 0.14 m	21		
		6.95 ± 0.07 m	2		
94	Rb	2.9 ± 0.3 s	22	2.72 ± 0.04 s	
		2.67 ± 0.04 s	23		
		2.8 ± 0.1 s	19		
		2.78 ± 0.05 s	2		
94	Sr	82 ± 4 s	22	75.6 ± 0.9 s	
		72 ± 7 s	24		
		84 ± 5 s	25		
		75.3 ± 0.7 s	2		
94	Y	20.3 ± 0.2 m	22	18.7 ± 1.2 m	The discrepancy between different measurements is large. No weight factors are used when calculating the average value
		19.4 ± 0.5 m	21		
		16.3 ± 0.2 m	2		
95	Sr	48 ± 9 s	22	28 ± 3 s	
		33 ± 6 s	25		
		26.8 ± 1.5 s	2		
95	Y	10.9 ± 0.2 m	22	10.6 ± 0.3 m	
		10.9 ± 0.1 m	25		
		10.2 ± 0.1 m	2		

Mass number	Element	Half-life Measured	Reference	Recommended	Comment
111	Ag	74 ± 3 s	26	65 ± 2 s	
		64.8 ± 0.8 s	2		
113	Ag	72 ± 9 s	27	69 ± 1 s	
		69 ± 1 s	2		
114	Ag	4.5 ± 0.3 s	28	4.35 ± 0.10 s	
		4.3 ± 0.1 s	2		
115	Ag	~ 20 s	27	18.0 ± 0.7 s	The half-life 55 s has also been assigned to ^{115}Ag [29, 30]
		18.0 ± 0.7 s	2		
116	Ag	10.4 ± 0.8 s	28	8.6 ± 0.4 s	
		8.5 ± 0.2 s	2		
116	Ag	150 ± 6 s	27	153 ± 4 s	
		155 ± 5 s	2		
117	Ag	5.34 ± 0.05 s	2		
117	Ag	66 ± 6 s	27	72.7 ± 0.8 s	
		$72.8^{+2.0}_{-0.7}$	2		
118	Ag	2.8 ± 0.3 s	28	2.8 ± 0.3 s	
118	Ag	3.7 ± 0.2 s	28	3.7 ± 0.2 s	
119	Cd	110 ± 10 s	31	110 ± 10 s	
119	Cd	160 ± 30 s	32	160 ± 10 s	
		160 ± 10 s	31		
119	In	170 ± 20 s	32	170 ± 20 s	
120	Ag	0.32 ± 0.04 s	28	0.32 ± 0.04 s	
120	Ag	1.17 ± 0.05 s	28	1.17 ± 0.05 s	
120	Cd	50.8 ± 0.2 s	33	50.8 ± 0.3 s	
		50.9 ± 0.5 s	2		
121	Ag	0.8 ± 0.1 s	34	0.8 ± 0.1 s	
121	Cd	7 ± 2 s	32,34	7 ± 2 s	
121	Cd	12.8 ± 0.4 s	35	13.6 ± 0.3 s	
		13 ± 2 s	32,34		
		13.8 ± 0.2 s	2		

Mass number	Element	Half-life Measured	Reference	Recommended	Comment
121	In	23 ± 2 s	34	23.1 ± 0.6 s	
		23.1 ± 0.6 s	2		
121	In	120 ± 40 s	34	120 ± 40 s	
122	Ag	1.5 ± 0.5 s	28	1.5 ± 0.5 s	
122	Cd	5.78 ± 0.09 s	33	5.78 ± 0.09 s	There is a great discrepancy between the two measurements. The value from ref. [33] is preferred because of less danger of interference from indium and silver.
		3.13 ± 0.12 s	2		
122	In	1.5 ± 0.3 s	36	1.5 ± 0.3 s	
122	In	7.5 ± 0.8 s	36	9.2 ± 0.3 s	
		9 ± 2	3		
		10.0 ± 0.5 s	37		
		9.2 ± 0.2 s	2		
123	Cd,In	10 ± 2 s	38	5.98 ± 0.06	A warning must be given here The 6 s activity could possibly be an isotope of cadmium. In that case the indium half-life should be 10±2 s.
		5.98 ± 0.06 s	2		
123	In	36 ± 3 s	38	48 ± 2 s	
		47.8 ± 0.5 s	2		
124	In	3.6 ± 1 s	39	3.17 ± 0.05 s	
		3.17 ± 0.05 s	2		
125	Cd,In	3.5 ± 1 s	3	2.33 ± 0.04 s	It is not known whether these activities are to be attributed to cadmium or indium
		2.33 ± 0.04 s	2		
125	Cd,In	12.2 ± 0.1 s	2	12.2 ± 0.1 s	
126	In	1.53 ± 0.01 s	2	1.53 ± 0.01 s	
127	In	1.09 ± 0.03 s	2	1.09 ± 0.03 s	
127	In	3.1 ± 0.3 s	2	3.1 ± 0.3 s	
127	Sn	276 ± 24 s	40	248 ± 2 s	
		246 ± 18 s	41		
		248 ± 2 s	2		

Mass number	Element	Half-life Measured	Reference	Half-life Recommended	Comment
128	In	0.80 ± 0.03 s	2	0.80 ± 0.03 s	
128	In	5.6 ± 0.4 s	2	5.6 ± 0.4 s	
129	In	0.8 ± 0.3 s	3	0.8 ± 0.3 s	
129	Sn	~ 120 s	42	135 ± 5 s	
		151 ± 7 s	43		
		134 ± 2 s	2		
129	Sn	8.8 ± 0.6 m	40	7.6 ± 0.2 m	
		7.5 ± 0.1 m	42		
		8.9 ± 0.6 m	2		
130	In	0.53 ± 0.05 s	44	0.53 ± 0.05 s	
130	Sn	7.1 ± 0.4 m	40	6.53 ± 0.10 m	
		6.51 ± 0.07 m	2		
131	In	0.3 ± 0.1 s	3	0.3 ± 0.1 s	
131	Sn	62.9 ± 2.5 s	43	61 ± 4 s	
		55 ± 4 s	2		
132	In	0.12 ± 0.02 s	45	0.12 ± 0.02 s	
132	Sn	41.1 ± 1.3 s	43	40.2 ± 0.5 s	
		40.6 ± 0.8 s	46		
		39.0 ± 1.0 s	47		
		40 ± 1 s	48		
		41.0 ± 1.5 s	2		
132	Sb	120 ± 10 s	49	185 ± 4 s	
		188 ± 20 s	50		
		162 ± 9 s	47		
		187 ± 1 s	47		
		173 ± 4 s	2		
133	Sn	1.7 ± 0.3 s	3	1.7 ± 0.3 s	
133	I	9 ± 2 s	3	9 ± 2 s	
134	Sb	0.85 ± 0.10 s	51	0.85 ± 0.10 s	
134	Sb	11.3 ± 0.3 s	52	10.7 ± 0.3 s	
		11.1 ± 0.8 s	53		
		10.3 ± 0.5 s	51		
		10.2 ± 0.3 s	2		

Mass number	Element	Half-life Measured	Reference	Half-life Recommended	Comment
135	Te	18 ± 2 s	54	19.2 ± 0.2 s	
		19.2 ± 0.2 s	2		
136	Te	20.9 ± 0.5 s	55	21.1 ± 0.7 s	
		24 ± 2 s	2		
136	I	48 ± 2	56	48 ± 2 s	Three isomeric states have been found in ^{136}I [56]
136	I	83 ± 3 s	56	83 ± 3 s	
136	I	100 ± 3 s	56	100 ± 3 s	
137	I	24.4 ± 0.4 s	17	24.6 ± 0.1 s	
		24.7 ± 0.1 s	16		
		24.5 ± 0.2 s	2		
137	Xe	234 ± 6 s	12	230 ± 2 s	
		229.1 ± 0.8 s	18		
		241 ± 3 s	2		
138	I	5.9 ± 0.4 s	12	6.6 ± 0.2 s	
		6.3 ± 0.7 s	17		
		6.62 ± 0.09 s	2		
139	I	2.7 ± 0.1 s	12	2.61 ± 0.10 s	
		2.0 ± 0.5 s	17		
		2.47 ± 0.15 s	2		
139	Xe	41.2 ± 0.15 s	57	40.4 ± 0.5 s	
		39.3 ± 0.7 s	58		
		39.68 ± 0.14 s	18		
		40. 8 ± 0.7 s	2		
140	Xe	13.7 ± 0.15 s	57	13.7 ± 0.2 s	
		14.3 ± 1.3 s	58		
		13.60 ± 0.10 s	18		
		15.4 ± 0.5 s	2		
140	Cs	63.8 ± 1.2 s	57	64.0 ± 0.4 s	
		65.7 ± 1.6 s	58		
		63.7 ± 0.3 s	18		
		65.5 ± 0.7 s	2		

Mass number	Element	Half-life Measured	Reference	Recommended	Comment
141	Xe	1.6 ± 0.1 s	59	1.72 ± 0.01 s	
		1.8 ± 0.2 s	58		
		1.720 ± 0.013 s	18		
		1.7 ± 0.2 s	2		
141	Cs	24.7 ± 0.4 s	18	25.2 ± 0.5 s	
		25.6 ± 0.3 s	2		
142	Cs	1.68 ± 0.02 s	18	1.79 ± 0.17 s	
		2.04 ± 0.03 s	2		
143	Ba	13.5 ± 0.3 s	60	13.6 ± 0.2 s	
		13.6 ± 0.2 s	2		
144	Ba	11.9 ± 0.3 s	60	10.8 ± 0.4 s	
		10.7 ± 0.2 s	2		
144	La	39.8 ± 0.6 s	61	40.3 ± 0.3 s	
		40.5 ± 0.4 s	2		
145	Ba	5.6 ± 0.6 s	61	4.8 ± 0.7 s	
		4.2 ± 0.5 s	2		
145	La	29.2 ± 0.8 s	61	29.3 ± 0.8 s	
		32 ± 4 s	2		
145	Ce	186 ± 12 s	62	192 ± 15 s	
		230 ± 30 s	2		

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Review Paper No. 13

STATUS OF DELAYED NEUTRON DATA

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Abstract:

Delayed neutron data are reviewed from the aspect of their use in reactor applications. Gross delayed neutron emission has been studied extensively in the past and available data appear to be sufficiently accurate. Problems encountered in measurement and evaluation of delayed neutron precursor data are discussed. Further work is required to obtain consistent sets of fission yields, delayed neutron yields and Pu values for individual precursors. Finally, recent high resolution results of delayed neutron energy spectra are presented.

I. INTRODUCTION

Delayed neutron data can be approached in two ways:

1. from the aspect of nuclear structure; this requires detailed and accurate information on the individual nuclides, as required in nuclear spectroscopy and reaction studies coupled to theoretical work. This is beyond the scope of this review.

2. from the aspect of the use, where the relevant data are of a required degree of detail and accuracy to meet the various needs and applications of delayed neutrons.

This is the purpose of the present paper, viz. to review and evaluate the available information on delayed neutron data from fission and point out problems which require further study.

The present topic served as a basis for a panel on "Delayed Fission Neutrons" organized, and its proceedings published, by IAEA in 1968. A number of compilations and reviews were prepared by Keepin[1], Herrmann et al.[2,3], Tomlinson[4,5], del Marmol[6], Pappas[7], Amiel[8,9] and others[10], the most recent (1972) ones being those of Tomlinson[4,11] and Schüssler and Herrmann[2].

Since this review will focus only on data aspects rather than on experimental and theoretical aspects, references are made to the sources used without making any attempt to exhaust all the important publications on the topic.

II. GROSS DELAYED NEUTRON EMISSION

Since delayed neutron emission is a phenomenon associated with the radioactive decay of a given number of fission produced nuclides, an accurate treatment of variations in abundance and gross properties should take into account cross sections and fission yields of the delayed neutron precursors in the fission reactions in question. The gross phenomenological observations of delayed neutrons are given in terms of delayed neutron yields, viz. the total number of delayed neutrons generated per fission in a given fission reaction. Since delayed neutron emission originates from a number of radionuclides decaying with various half-lives, the separation of the time dependent neutron emission results in a number of half-life groups. Due to the similarity of the mass distributions in fission from various sources,

TABLE 1: Absolute delayed neutron yields (neutrons per 10^4 fissions)

Fission nuclide	Neutron energy				
	Thermal	Fission Spect.	0.1-1.8 MeV	3.1 MeV	~15 MeV
^{232}Th	-	496 ± 35[a]	-	570 ± 50[d]	300 ± 20[d] 190 ± 30[f]
^{233}U	66 ± 3[a]	70 ± 6[a]	75 ± 6[b]	74 ± 6[d]	41 ± 3[d]
^{235}U	158 ± 5[a]	165 ± 7[a]	163 ± 13[b]	172 ± 13[d]	91 ± 4[d]
^{238}U	-	412 ± 25[a]	-	484 ± 36[d]	283 ± 13[d] 230 ± 80[f]
^{239}Pu	61 ± 3[a]	63 ± 5[a]	62 ± 5[b]	66 ± 5[d]	41 ± 2[d]
^{240}Pu		88 ± 6[a]			
^{241}Pu	159 ± 16[e]				
^{242}Pu			150 ± 50[b,c]		

a. Keepin[1]

b. Krick and Evans[31], see revision in Evans, Thorpe and Krick[13]

c. averaged 0.6 to 1.3 MeV

d. Masters, Thorpe and Smith[12], see revision in Evans, Thorpe and Krick[13]

e. Cox, corrected by Tomlinson[4]

f. Benedict, Luthardt, Hermann[32]

there exists a general similarity in half-lives and abundances of the respective d.n. emission groups from various sources.

The multicomponent decay curve of the delayed neutron emission from an irradiated fissionable source was shown by Keepin[1] to be well resolved into six exponential components. Of course a greater number of exponentials will result in a more realistic description, but six exponentials serve as a satisfactory optimum for most practical purposes.

Table 1 presents the absolute delayed neutron yields of various fission sources. Table 2 and Fig. 1 give the variation of the delayed neutron yield with energy.

TABLE 2: Delayed-neutron yield data at various fission inducing neutron energies*

^{235}U		^{233}U		^{239}Pu	^{238}U	
E_n	Yield, n/fission	E_n	Yield, n/fission		E_n	Yield, n/fission[a]
0.05	0.0165 ± 0.0013	0.05	0.00735 ± 0.00064	0.00610 ± 0.0005	1.65	0.0484 ± 0.0040
0.15	0.0162 ± 0.0012	0.11	0.00735 ± 0.00064	0.00602 ± 0.0005	4.00	0.0484 ± 0.0045
0.25	0.0166 ± 0.0013	0.21	0.00730 ± 0.00064	0.00602 ± 0.0005	4.25	0.0483 ± 0.0044
0.36	0.0165 ± 0.0013	0.32	0.00741 ± 0.00064	0.00610 ± 0.0005	4.50	0.0474 ± 0.0043
0.46	0.0162 ± 0.0012	0.43	0.00734 ± 0.00064	0.00615 ± 0.0005	4.75	0.0482 ± 0.0041
0.56	0.0160 ± 0.0012	0.53	0.00745 ± 0.00065	0.00626 ± 0.0005	5.15	0.0464 ± 0.0043
0.67	0.0160 ± 0.0012	0.64	0.00745 ± 0.00065	0.00631 ± 0.0005	5.35	0.0453 ± 0.0043
0.77	0.0162 ± 0.0012	0.74	0.00759 ± 0.00066	0.00633 ± 0.0005	5.50	0.0429 ± 0.0041
0.87	0.0163 ± 0.0013	0.84	0.00759 ± 0.00066	0.00624 ± 0.0005	5.75	0.0424 ± 0.0040
0.97	0.0160 ± 0.0012	0.94	0.00719 ± 0.00065	0.00621 ± 0.0005	6.00	0.0412 ± 0.0041
1.07	0.0163 ± 0.0013	1.05	0.00759 ± 0.00066	0.00626 ± 0.0005	6.30	0.0404 ± 0.0039
1.17	0.0167 ± 0.0013	1.15	0.00759 ± 0.00066	0.00626 ± 0.0005	6.50	0.0395 ± 0.0039
1.27	0.0171 ± 0.0013	1.25	0.00741 ± 0.00065	0.00629 ± 0.0005	6.70	0.0381 ± 0.0038
1.37	0.0170 ± 0.0013	1.35	0.00765 ± 0.00066	0.00633 ± 0.0005	6.90	0.0374 ± 0.0039
1.47	0.0160 ± 0.0012	1.45	0.00765 ± 0.00066	0.00633 ± 0.0005		
1.56	0.0167 ± 0.0013	1.55	0.00759 ± 0.00066	0.00624 ± 0.0005	^{242}Pu	
1.65	0.0163 ± 0.0013	1.65	0.00739 ± 0.00064	0.00621 ± 0.0005	0.64	0.0151 ± 0.0045
1.75	0.0161 ± 0.0012	1.75	0.00759 ± 0.00066	0.00626 ± 0.0005	0.84	0.0149 ± 0.0045
4.0	0.0153 ± 0.0015	4.0	0.0080 ± 0.0009		1.05	0.0153 ± 0.0045
4.4	0.0146 ± 0.0015	4.5	0.0074 ± 0.0009		1.25	0.0160 ± 0.0045
4.8	0.0151 ± 0.0014	5.1	0.0070 ± 0.0008			
5.1	0.0136 ± 0.0014	5.35	0.0065 ± 0.0008			
5.5	0.0125 ± 0.0013	5.6	0.0055 ± 0.0007			
5.7	0.0111 ± 0.0013	6.1	0.0050 ± 0.0006			
6.0	0.0123 ± 0.0013	6.6	0.0051 ± 0.0006			
6.4	0.0102 ± 0.0012					
6.7	0.0105 ± 0.0012					

* Data produced by Krick and Evans[31], as given by Evans, Thorpe and Krick[13]

a. Absolute values based on normalization to 3.1 MeV data of Masters, Thorpe and Smith[12]

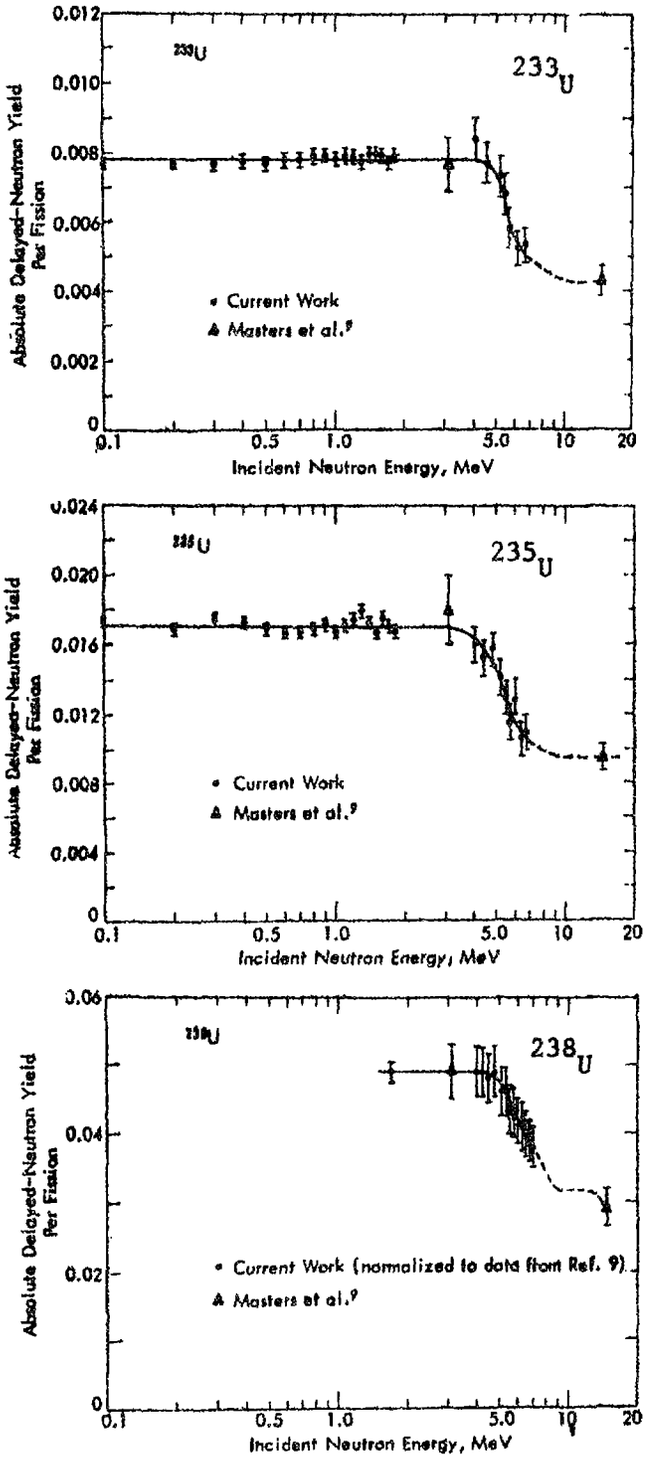

Fig. 1. Delayed neutron yields as a function of energy for various uranium isotopes. (Taken from Ref. 31)
Current work: Krick and Evans[31], 9) Masters, Thorpe and Smith[12]

It seems that the existing programs (especially at Los Alamos[12,13] Purdue[14] and Argonne[15]) did result in sufficiently accurate data for the delayed neutron yields of the commonly useful fissionable nuclides in a range of energies from thermal neutrons to ~5 MeV and up to ~15 MeV. The constancy of the d.n.y. over a wide range of energies, up to ~4.5 MeV is an expected phenomenon, while the sharp drop in the d.n. yield can be well explained by neutron evaporation from the compound nucleus prior to fission, which takes place (at the "second chance") from a one neutron higher nuclide. A systematic and quantitative account of this effect has been made by Cox[15].

TABLE 3a: Group half-lives and yields: Thermal fission†

Group	half-life (sec)	λ (sec^{-1})	relative abundance	absolute group yield (n/100F)
		^{233}U		
1	55.00 0.80	0.0126 0.0004	0.086 0.004	0.060 0.005
2	20.57 0.56	0.0337 0.0009	0.299 0.006	0.206 0.013
3	5.00 0.31	0.139 0.009	0.252 0.059	0.173 0.040
4	2.13 0.30	0.325 0.045	0.278 0.030	0.192 0.024
5	0.615 0.359	1.13 0.60	0.051 0.036	0.036 0.024
6	0.277 0.070	2.50 0.62	0.034 0.021	0.023 0.013
		^{235}U		
1	55.72 1.90	0.0124 0.0004	0.033 0.004	0.054 0.007
2	22.72 1.05	0.0305 0.0015	0.219 0.013	0.362 0.027
3	6.22 0.34	0.111 0.006	0.196 0.033	0.324 0.053
4	2.30 0.13	0.301 0.016	0.395 0.016	0.651 0.039
5	0.610 0.123	1.14 0.22	0.115 0.013	0.190 0.022
6	0.230 0.037	3.01 0.43	0.042 0.012	0.069 0.012
		^{239}Pu		
1	54.28 3.47	0.0128 0.0007	0.035 0.013	0.022 0.009
2	23.04 2.48	0.0301 0.0033	0.298 0.052	0.191 0.034
3	5.60 0.59	0.124 0.013	0.211 0.071	0.135 0.045
4	2.13 0.36	0.325 0.053	0.326 0.049	0.209 0.033
5	0.618 0.316	1.12 0.58	0.086 0.043	0.055 0.027
6	0.257 0.067	2.69 0.71	0.044 0.024	0.028 0.015
		^{241}Pu		
1	54.0 1.0	0.0128 0.0002	0.010 0.003	0.0154 0.004
2	23.2 0.5	0.0299 0.011	0.229 0.006	0.365 0.010
3	5.6 0.6	0.124 0.013	0.173 0.025	0.275 0.040
4	1.97 0.1	0.352 0.018	0.390 0.050	0.620 0.080
5	0.43 0.04	1.61 0.15	0.182 0.019	0.290 0.030
6	0.2+ 0.1+	3.47+ 1.7+	0.016*0.006+	0.026* 0.01+

(Errors are 1σ. Data for ^{233}U, ^{235}U and ^{239}Pu are taken from Keepin et al.[1]. Data for ^{241}Pu are from Cox)

† prepared by Tomlinson[4]

+,* estimated

Table 3a, b (taken from Ref. 4) presents the variously analyzed d.n. groups in thermal and fast neutron fission of the common fissionable nuclides. Results with lesser precision were also obtained for ^{231}Pa [16], ^{237}Np and ^{241}Am [17] and ^{252}Cf [18].

Figure 2 presents the observed mass distribution (at low resolution) of delayed neutron emission in fission of ^{235}U (from Ref. 19).

TABLE 3b: Group half-lives and yields: Fast fission (fission spectrum)*

Group	half-life (sec)		λ_{-1} (sec^{-1})		relative abundance		absolute group yield (n/100F)	
			^{232}Th					
1	56.03	1.41	0.0124	0.0003	0.034	0.003	0.177	0.018
2	20.75	0.98	0.0334	0.0016	0.150	0.007	0.780	0.055
3	5.74	0.36	0.121	0.007	0.155	0.031	0.806	0.160
4	2.16	0.12	0.321	0.016	0.446	0.022	2.319	0.163
5	0.571	0.062	1.21	0.13	0.172	0.019	0.895	0.108
6	0.211	0.028	3.29	0.441	0.043	0.009	0.223	0.046
			^{233}U					
1	55.11	2.76	0.0126	0.0006	0.086	0.004	0.059	0.004
2	20.74	1.28	0.0334	0.0021	0.274	0.007	0.189	0.013
3	5.30	0.28	0.131	0.007	0.227	0.052	0.157	0.037
4	2.29	0.27	0.302	0.036	0.317	0.016	0.219	0.018
5	0.546	0.160	1.27	0.395	0.073	0.021	0.050	0.015
6	0.221	0.062	3.13	1.00	0.023	0.010	0.016	0.007
			^{235}U					
1	54.51	1.39	0.0127	0.0003	0.038	0.004	0.083	0.007
2	21.84	0.80	0.0317	0.0012	0.213	0.007	0.351	0.016
3	6.00	0.25	0.115	0.004	0.188	0.024	0.310	0.042
4	2.23	0.09	0.311	0.012	0.407	0.010	0.672	0.034
5	0.496	0.043	1.40	0.012	0.128	0.012	0.211	0.022
6	0.179	0.025	3.87	0.546	0.026	0.004	0.043	0.007
			^{238}U					
1	52.38	1.91	0.0132	0.0004	0.013	0.001	0.058	0.007
2	21.58	0.58	0.0321	0.0009	0.137	0.003	0.602	0.037
3	5.00	0.28	0.139	0.007	0.162	0.030	0.712	0.129
4	1.93	0.10	0.358	0.021	0.388	0.018	1.708	0.120
5	0.493	0.034	1.41	0.099	0.225	0.019	0.989	0.089
6	0.172	0.013	4.02	0.317	0.075	0.007	0.330	0.036

(continued)

* prepared by Tomlinson [4]

TABLE 3b (continued):

Group	half-life (sec)	λ_{-1} (sec^{-1})	relative abundance	absolute group yield (n/100F)
		^{239}Pu		
1	53.75 1.41	0.0129 0.0003	0.038 0.004	0.024 0.003
2	22.29 0.53	0.0311 0.0007	0.280 0.006	0.179 0.013
3	5.19 0.18	0.134 0.004	0.216 0.027	0.138 0.019
4	2.09 0.12	0.331 0.018	0.328 0.015	0.210 0.018
5	0.549 0.073	1.26 0.171	0.103 0.013	0.066 0.010
6	0.216 0.025	3.21 0.378	0.035 0.007	0.022 0.004
		^{240}Pu		
1	53.56 1.80	0.0129 0.0006	0.028 0.004	0.022 0.004
2	22.14 0.56	0.0313 0.0007	0.273 0.006	0.238 0.024
3	5.14 0.62	0.135 0.016	0.192 0.079	0.162 0.065
4	2.08 0.28	0.333 0.046	0.350 0.030	0.315 0.040
5	0.511 0.114	1.36 0.304	0.128 0.027	0.119 0.027
6	0.172 0.049	4.04 1.16	0.029 0.009	0.024 0.007
		^{242}Pu (estimated values)		
1	53.8	0.0129	0.004	0.006
2	23.5	0.0295	0.195	0.31
3	5.3	0.131	0.162	0.26
4	2.05	0.338	0.411	0.66
5	0.50	1.39	0.218	0.35
6	0.19	3.65	0.010	0.016

(Errors are 1σ. Data for all nuclides except ^{242}Pu are taken from Keepin et al. [1]. All data for ^{242}Pu are estimated. The relative yields are from Cox et al. ; the half-lives are averages of group half-lives for Pu isotopes; the absolute group yields are estimated from the total delayed neutron yield given in Krick and Evans [31] and the relative yields in Cox et al.)

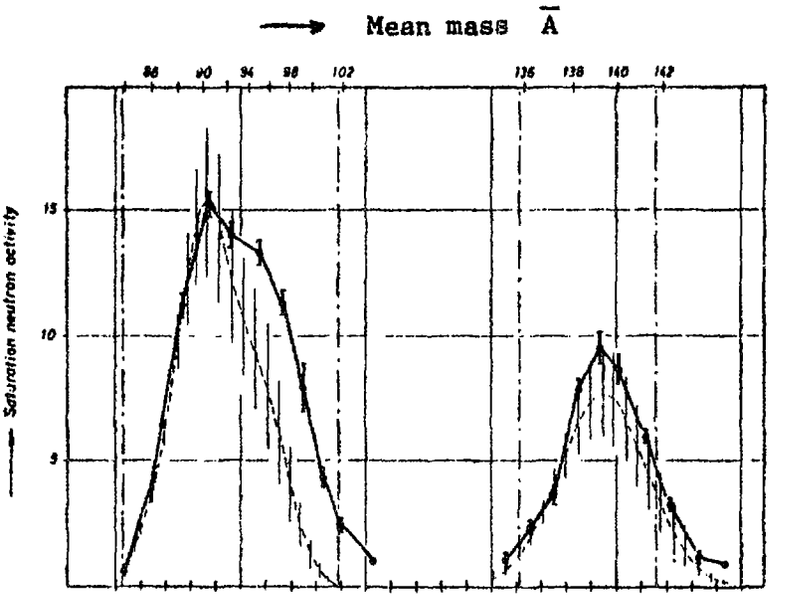

Fig. 2. Mass dependence of saturation neutron activity (determined by neutron counting of mass separated fission fragments). From Ref.19.

III. DELAYED NEUTRON PRECURSORS

The available data on the known delayed neutron precursors are listed in Table 4. In Table 5 an attempt has been made to compare the contributions of the individual precursors to the d.n. groups and total delayed neutron emission in low energy fission of ^{235}U. The fit seems satisfactory, but there are a few basic problems that need further clarification. The d.n. yield is a product of the cumulative fission yield and the Pn value. Only several Pn values were measured directly with satisfactory accuracy. This requires good isotopic purity, which can be obtained by techniques introduced only recently. Most of the directly measured values are the d.n. yields. This requires good chemical purity and sufficient resolution of the various decaying components, to allow correct assignment of the neutron yield. The question then remains regarding the fission yields. The accuracy of such data is open to argument. In a recent paper[20] a detailed analysis of data revealed a systematically pronounced odd-even effect ($\pm$25%) which implies a major revision of the fission yield used in many calculations.

TABLE 4: Identified delayed neutron precursors and neutron emission probabilities

Precursor (and half-life)		Cumulated fission yield ^{235}U-th.fiss.(a)	Delayed neutron yield		Pn(%)			
			Experimental	Calculated	Calculated values			Experimental
					This work	Mains[22]	Tomlinson[11] **	
5.6s	^{84}As	0.33±0.05*	0.02±0.01[33]		0.06±0.03	0.13±0.06		
2.0s	^{85}As	0.13±0.07	7.8 ±1.2[33]		60±34	23±3		
0.9s	^{86}As	0.08±0.02	0.57±0.2[33]		7.1±3.0	3.8±1.0		
5.6s	^{87}Se	0.9±0.2		0.22±0.07[35] 0.15±0.04[36]		0.25±0.06		0.25±0.06[35]+ 0.16±0.03[36]+++
1.5s	^{88}Se	0.39±0.04		0.06±0.04[35] 0.29±0.04[36]		0.15±0.09		0.15±0.09[35]++ 0.75±0.06[36]+++
0.4s	^{89}Se	0.11±0.05		0.55±0.30[36]				5.0 ±1.5[36]+++
55.7s	^{87}Br	2.02±0.2	5.1±0.8[2],	4.24±0.74[37] 6.26±1.36[38]	2.5±0.44	2.3±0.4		3.1±0.6[38] 2.1±0.3[37]
16s	^{88}Br	2.2±0.2	12.1±1.2[2]	12.3±28[11] 13.2±3.7[38]	5.5±0.74	4.2±0.5	5.6±1.2	6.0±1.6[38]
4.5s	^{89}Br	1.36±0.13	17±3[2]	16.7±3.6[11] 9.5±2.8[38]	12.5±2.5	6.2±1.3	12.3±26	7±2[38]
1.6s	^{90}Br	0.65±0.03	16±3[2]	10.4±4.2[11]	24.6±4.75	7.8±2.2	16±6.5	
0.6s	^{91}Br	0.23±0.02	2.9±1.4[2]		12.6±6.2	8.4±3.3		
0.25s	^{92}Br	∿0.035				16^{+16}_{-8}		
1.8s	^{92}Kr	1.86±0.1		0.074±0.013[39]				0.04±0.007[39]
1.3s	^{93}Kr	0.54±0.05		1.40±0.30[39]				2.60±0.50[39]
0.2s	^{94}Kr	0.17±0.02						
4.5s	^{92}Rb	5.06±0.3		0.06±0.02[39]				0.012±0.004[39]
5.9s	^{93}Rb	3.64±0.2	6.3±1.8[2]	5.20±0.70[40] 6.00±1.14[39]	1.73±0.50			1.43±0.18[40] 1.65±0.30[39]
2.7s	^{94}Rb	1.8±0.2	16.7±2.6[2]	15.2±2.3[41]	9.3±1.8			8.46±0.92[41]
0.38s	^{95}Rb	0.66±0.05		5.64±0.74[41]				8.54±0.91[41]
0.2s	^{96}Rb	∿0.014		∿1.82±0.20[41]				13±1.4[41]
0.17s	^{97}Rb	∿0.035		∿0.95±0.10[41]				27.2±3.0[41]
0.11s	^{98}Rb							13.3±2.1[41]
1.1s	^{97}Y	4.85 0.5						
<0.3s	^{98}Y	3.14±0.3	9.2±4[2]			4±2		
∿0.8s	^{99}Y	1.9±0.2						
11s	^{134}Sb	0.32±0.03	0.028±0.004[34]	0.025±0.003[11]	∿0.094±0.010		0.08	
1.7s	^{135}Sb	0.13±0.03	3.5±0.3[34]	1.04±0.26[11]	27±6.6		8±2	
21s	^{136}Te	1.60±0.2	∿1.2[3]		∿0.75±0.10	∿0.5		
3.5s	^{137}Te	0.5±0.2	∿0.7[3]		∿1.4±0.3	∿0.5		
24.6s	^{137}I	3.27±0.5	21.7±1.9[2]	9.81±2.22[38] 28.1±5.6[37]	6.65±1.17	5.2±0.7		3.0±0.5[38] 8.6±1.2[37]
6.5s	^{138}I	1.4±0.5	7.2±1.5[2]	2.80±1.30[38]	5.14±2.12	3.0±0.6		2.0±0.6[38]
2.6s	^{139}I	0.67±0.06	9.3±1.6[2]		13.9±2.7	6.5±1.7		
0.86s	^{140}I	0.13±0.02	6.5±2.5[2]		50±20	14±5		
0.44s	^{141}I	∿0.017						
1.7s	^{141}Xe	1.2±0.12		0.064±0.012[39]				0.054±0.009[39]
1.2s	^{142}Xe	0.63±0.1		0.28±0.07[39]				0.45±0.08[39]
25s	^{141}Cs	4.3±0.3		0.314±0.052[39]				0.073[39]
1.9s	^{142}Cs	3.0±0.3		0.81±0.22[39]				0.27±0.07[39]
1.7s	^{143}Cs	1.5±0.2		1.69±0.43[40]				1.13±0.25[40]
1.1s	^{144}Cs	0.3±0.1		0.33±0.13[40]				1.10±0.25[40]
0.61s	^{145}Cs	0.1±0.05		1.2±0.6[41]				12.1±1.4[41]
0.35s	^{146}Cs							14.2±1.7[41]

* According to Ref. [43] the cumulative yield was measured to be half of the quoted value, i.e. 0.16, which leads to Pn=0.12. The yield measured by the Mains group[44] is in agreement with the value given in the table.

** Calculated by Tomlinson from relative yields obtained from Ref. [42]

\+ Based on Pn(^{87}Br) = 2.4

++ Based on Pn(^{88}Br) = 4.0

+++ Normalized by the author to Pn values of 2.3, 4.7, 5 for ^{87}Br, ^{88}Br, ^{89}Br, respectively.

(a) Based on the odd-even systematics in the fission yields[20]. As and Se values are of lower accuracy due to scarcity of data and conflicting published results.

TABLE 5: Summary of the contributions of identified precursors to delayed neutron group yields in thermal neutron fission of ^{235}U

	Group	Group half-life	Precursor		Delayed Neutron yield (n/10^4fiss)*		Unseparated sample(1)
	1	55s	56s	^{37}Br	5.1	± 0.8	
subtotal					5.1	± 0.8	5.2 ± 0.5
	2	22s	16s	^{88}Br	12.1	± 1.2	
			21s	^{136}Te	∿ 1.2		
			25s	^{137}I	21.7	± 1.9	
			25s	^{141}Cs	[0.31]		
subtotal					35.3	± 2.3	34.6 ± 1.8
	2-3		11s	^{134}Sb	0.028		
	3	6s	5.6s	^{84}As	0.02	± 0.01	
			5.6s	^{87}Se	[0.18]		
			4.5s	^{89}Br	17	± 3	
			4.5s	^{92}Rb	[0.06]		
			5.9s	^{93}Rb	6.3	± 1.8	
			3.5s	^{137}Te	∿ 0.7		
			6.5s	^{138}I	7.2	± 1.5	
subtotal					31.46	± 3.8	31.0 ± 3.6
	4	2s	2.0s	^{85}As	7.8	± 1.2	
			1.5s	^{88}Se	[0.2]	± 0.1	
			1.6s	^{90}Br	16	± 3	
			1.8s	^{92}Kr	0.074		
			1.3s	^{93}Kr	[1.4]	± 0.3	
			2.7s	^{94}Rb	16.7	± 2.6	
			1.7s	^{135}Sb	3.5	± 0.3	
			2.6s	^{139}I	9.3	± 1.6	
			1.7s	^{141}Xe	0.064		
			1.2s	^{142}Xe	[0.28]		
			1.9s	^{142}Cs	[0.81]		
			1.7s	^{143}Cs	[1.69]		
			1.1s	^{144}Cs	[0.33]		
subtotal					58.15	± 4.5	62.4 ± 2.6

./...

* Calculated yields are given in brackets; others are experimental values from Table 4.

TABLE 5 (contd)

	Group	Group Half-life	Percursor	Delayed Neutron yield (n/10^4fiss)*	Unseparated sample(1)
	5-6	0.6-0.2s	0.9s ^{36}As	0.57 ± 0.2	
			0.4s ^{89}Se	[0.55]	
			0.6s ^{91}Br	2.9 ± 1.4	
			0.25s ^{92}Br	[0.57]	
			0.4s ^{95}Rb	[5.64 ± 0.74]	
			0.2s ^{96}Rb	[∿1.8]	
			0.2s ^{97}Rb	[∿0.95]	
			$^{98,99}Y$	9 ± 4	
			0.9s ^{140}I	6.5 ± 2.5	
			0.4s ^{141}I	[0.5]	
			0.6s ^{145}Cs	[1.2 ± 0.6]	
			0.3s ^{146}Cs	[low]	
subtotal				30.2 ± 5.0	24.8 ± 2.3
TOTAL d.n.y.				160 ± 8	158 ± 5

* Calculated yields are given in brackets; others are experimental values from Table 4.

Such a treatment was introduced in Table 4. To extend this treatment of calculating Pn values for measured d.n. yields, or conversely, calculating d.n. yields from measured Pn values for fission reactions other than of ^{235}U, requires examination of the fine structure of the fission yield values in the corresponding nuclides. This information is not yet available with satisfactory accuracy.

For example, from Ref. 18 it is known that delayed neutron precursors, other than those mentioned in Table 4, do contribute significantly (~35%) to the d.n. emission in ^{252}Cf (precursors of masses 105-115, 140-150). A full account requires the identification of these species, then measurement of either their Pn values or their d.n. yields. This, together with the knowledge of the systematics of the fission yields, will permit a more complete account of d.n. precursors and calculation of their contributions to the entire range of fission reactions between thorium and the very heavy actinides.

TABLE 5a : DELAYED NEUTRON YIELDS IN THERMAL FISSION OF ^{235}U ; DISTRIBUTION OF YIELDS

a) ACCORDING ISOTOPES, ELEMENTS AND MASSES

	A	As	Se	Br	Kr	Rb	Y	(d.n.y)A	Peak, Total
		(8.39)	*(0.93)*	*(53.1)*	*(1.47)*	*(31.45)*	*(9)*		
	84	0.02						0.02	
	85	7.8						7.80	
	86	0.57						0.57	
	87		0.18	5.1				5.28	
	88		0.2	12.1				12.30	
	89		0.55	17				17.55	
	90			16				16.00	
	91			2.9				2.90	
LIGHT	92				0.074	0.06		0.704	105.09
PEAK	93				1.4	6.3		7.70	(65.5%)
	94					16.7		16.70	
	95					5.64		5.64	
	96					1.8		1.80	
	97					0.95		0.95	
	98						9	9.00	
	99								
		Sb	Te	I	Xe	Cs			160.4
		(3.53)	*(1.9)*	*(45.2)*	*(0.344)*	*(4.34)*			
	134	0.03						0.03	
	135	3.5						3.5	
	136		1.2					1.2	
	137		0.7	21.7				22.4	
	138			7.2				7.2	
HEAVY	139			9.3				9.3	55.3
PEAK	140			6.5				6.5	34.5%
	141			0.5	0.064	0.31		0.874	
	142				0.28	0.81		1.09	
	143					1.69		1.69	
	144					0.33		0.33	
	145					1.2		1.2	
	146					low		low	

b) ACCORDING CHEMICAL GROUPS

		d.n.y.		% of total d.n.y.
		$n/10^4 f$	%	
Halogens:	Br	53.1	33.1%	61.3%
	I	45.2	28.2%	
Alkalis:	Rb	31.45	19.6%	22.3%
	Cs	4.34	2.7%	
Noble Gases:	Kr	1.47	0.92%	1.13%
	Xe	0.34	0.21%	
V Group:	As	8.39	5.2%	7.4%
	Sb	3.5	2.2%	
VI Group:	Se	0.93	0.57%	1.75%
	Te	1.9	1.18%	
Rest:		~9	5.6%	5.6%

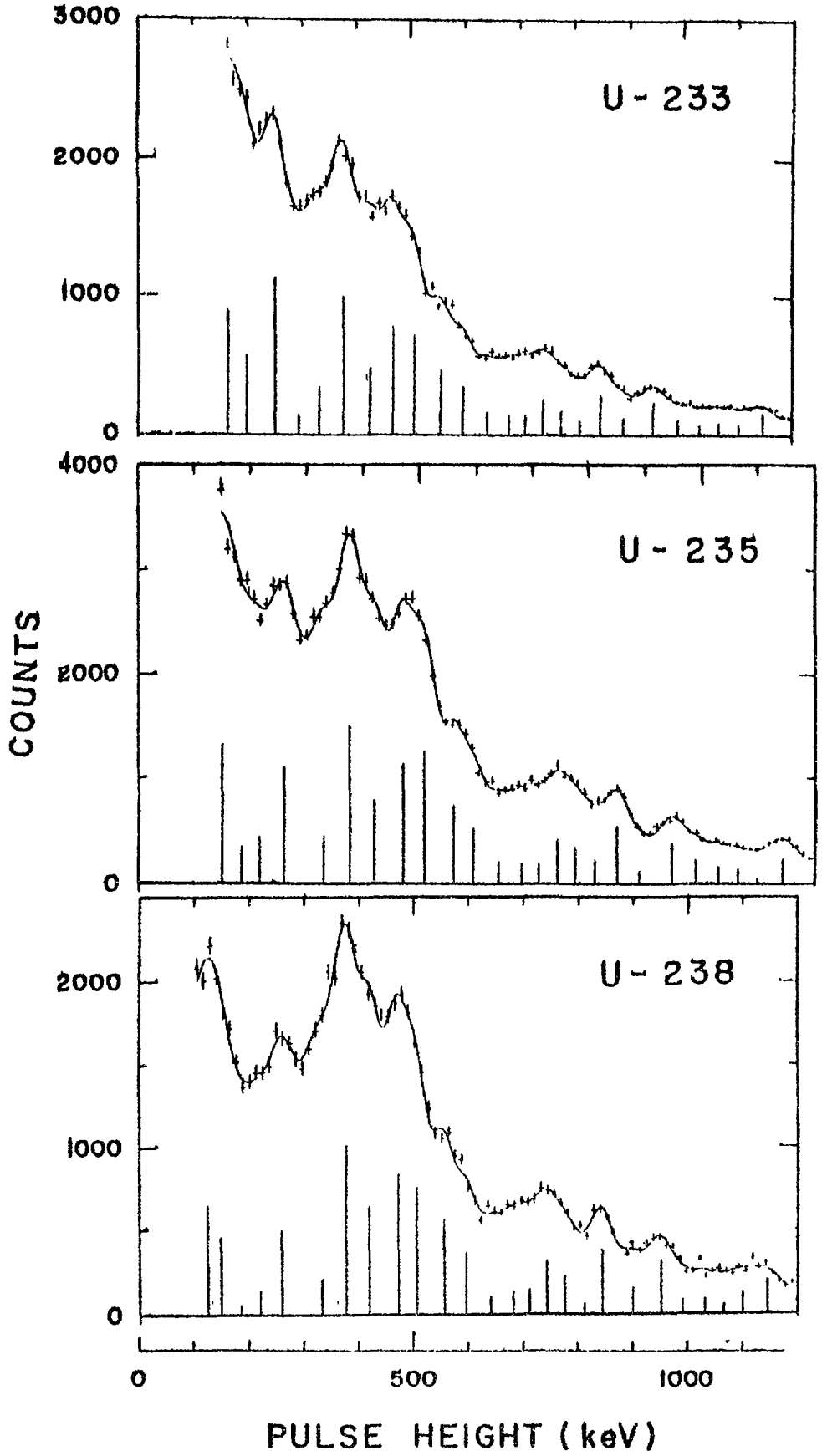

Fig. 3. Delayed neutron spectra (from Shalev and Cuttler, Ref. 25)

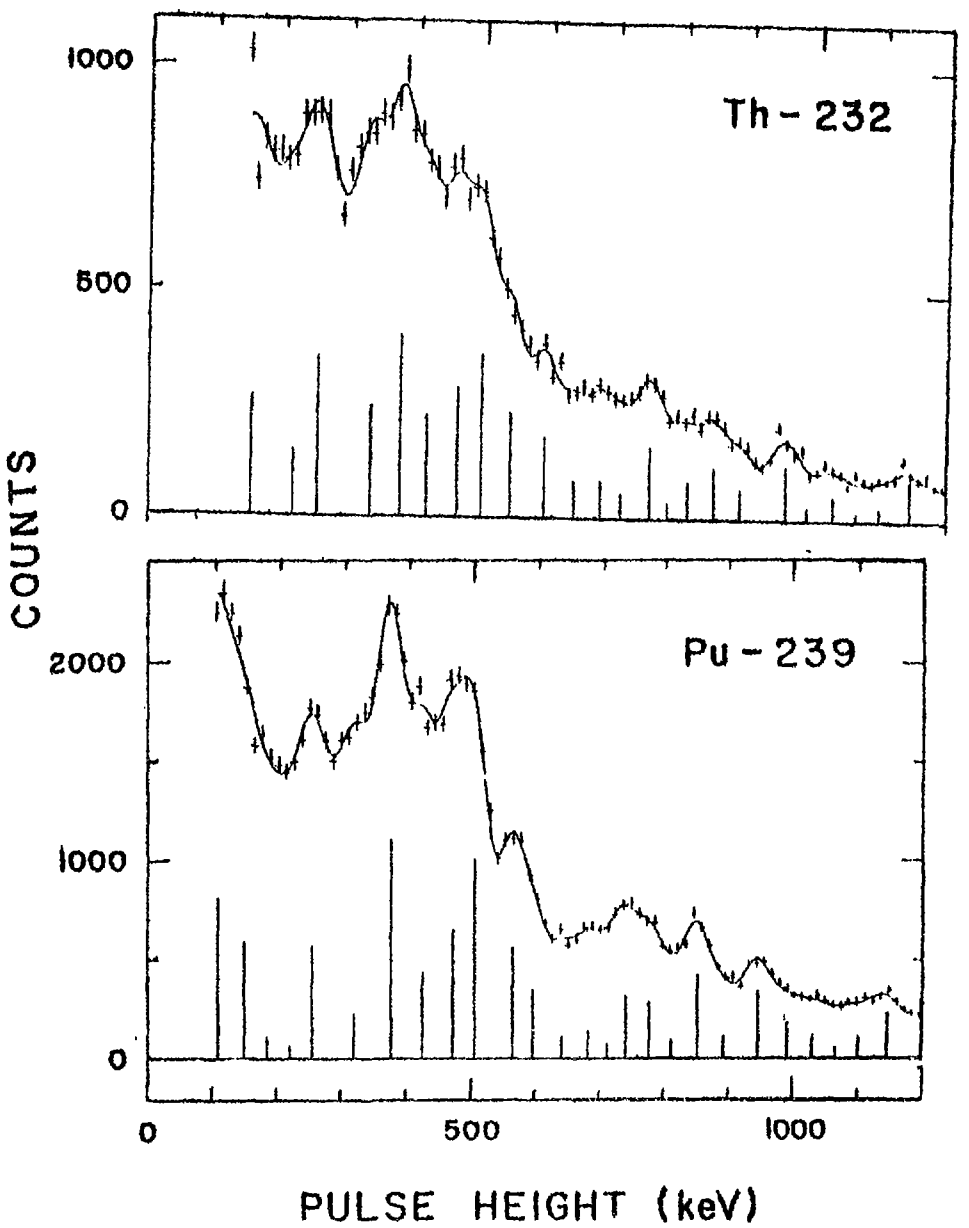

Fig. 3 continued

Attempts to predict Pn values on the grounds of nuclear parameters were reported by several authors[21-24], but fits of several tens of percents with data (masses, binding energies and Pn values) which are of the same accuracy are too coarse to draw conclusions on the accurate theoretical treatment at this stage.

IV. DELAYED NEUTRON ENERGY SPECTRA

The recent introduction of high resolution ^{3}He spectrometry by Shalev et al.[25] permits studies of detailed structures of both gross d.n. spectra (Fig. 3) [25] and spectra of individual precursors (Fig. 4) [26].

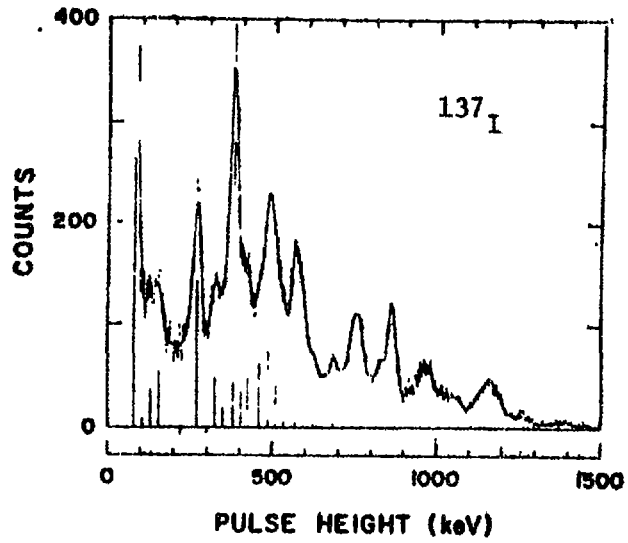

Fig. 4. Experimental pulse-height distribution of delayed neutrons from ^{137}Xe. (Taken from Ref.26)

TABLE 6. Mean energies of group and steady-state spectra, relative error of the values ±10 per cent*

Measurement	Mean energies (keV) group 1	2	3	4	5	Steady-state spectra
^{235}U (therm.)	277	484	447	432	—	435
^{238}U (14 MeV)	286	458	432	480	—	451
^{235}U (14 MeV)	278	468	443	425	382	445
^{239}Pu (14 MeV)	296	481	411	430	—	425
BATCHELOR and HYDER (1956) (^{235}U therm.)	250 ± 20	460 ± 10	405 ± 20	450 ± 20	—	430
BURGY *et al.* (1946) (^{235}U therm.)	300 ± 60	670 ± 10	650 ± 90	910 ± 90	400 ± 70	

* Taken from Fieg, Ref. 27.

Other studies of well-resolved gross neutron spectra from fission of ^{235}U and ^{239}Pu were reported by Fieg[27] with results similar to Fig. 3, as shown in Table 6. Sloan and Woodruff[28] report of a substantial number of resolved peaks in the ^{235}U d.n. spectra, as shown in Table 7, and compare them with results obtained by other groups.

A great number of neutron spectra of isotopically separated precursors are being processed presently by Rudstam and Shalev and are expected to be published soon* (Fig. 4 being an early study). An

* Published these Panel proceedings, Vol. 3 .

TABLE 7. Peak locations (taken from Ref.28)

Sloan and Woodruff[28] (keV)	Chulick et al.[45] (keV)	Chrysochoides et al[16] (keV)	Shalev[25] (keV)
33 ± 1			
41 ± 1		45 ± 2	
60 ± 3		[65, 70][3]	
76 ± 3		80 ± 5	75
90 ± 2	95 ± 4		
103 ± 4	104 ± 4	105 ± 6	
129 ± 5	135 ± 5	145 ± 13	125
161 ± 4	158 ± 8		170
183 ± 9	188 ± 10		180*
210(1)	205 ± 11		
	225 ± 13		
240 ± 5	240 ± 18		255*
283 ± 7	265 ± 17	260	
351 ± 9	340 ± 25		[320, 390][4]
420 ± 8	430 ± 32		420*
467 ± 11			440
			480*
			500
552 ± 20			570*
(see footnote 2)			680*
			750*
			850*
			950*
			1145*

* Shalev long cycle data

[1]Some caution is warranted for the region 200 keV to 220 keV due to the overlapping of the methane and the hydrogen detectors.

[2]Statistical quality and resolution of the data above 600 keV did not permit easy identification of peaks and are not reported. There is, however, qualitative agreement with the Shalev data.

[3]Reference 16 indicated a peak somewhere in this interval.

[4]Shalev reports peaks at 320, 325, 335, 375 and 390 keV in this interval although this implies resolution better than that reported. Reference 25a indicates that some caution should be exercised with respect to peak locations previously tabulated in Reference 25.

example of a very fine study of ^{85}As by the Mainz group[29] is seen in Fig. 5[30] and Fig. 6[29]. In the particular case of ^{85}As the entire spectrum, of both the electromagnetic and neutron transitions, is fully accounted for. This is also the first case where most neutrons populate excited states.

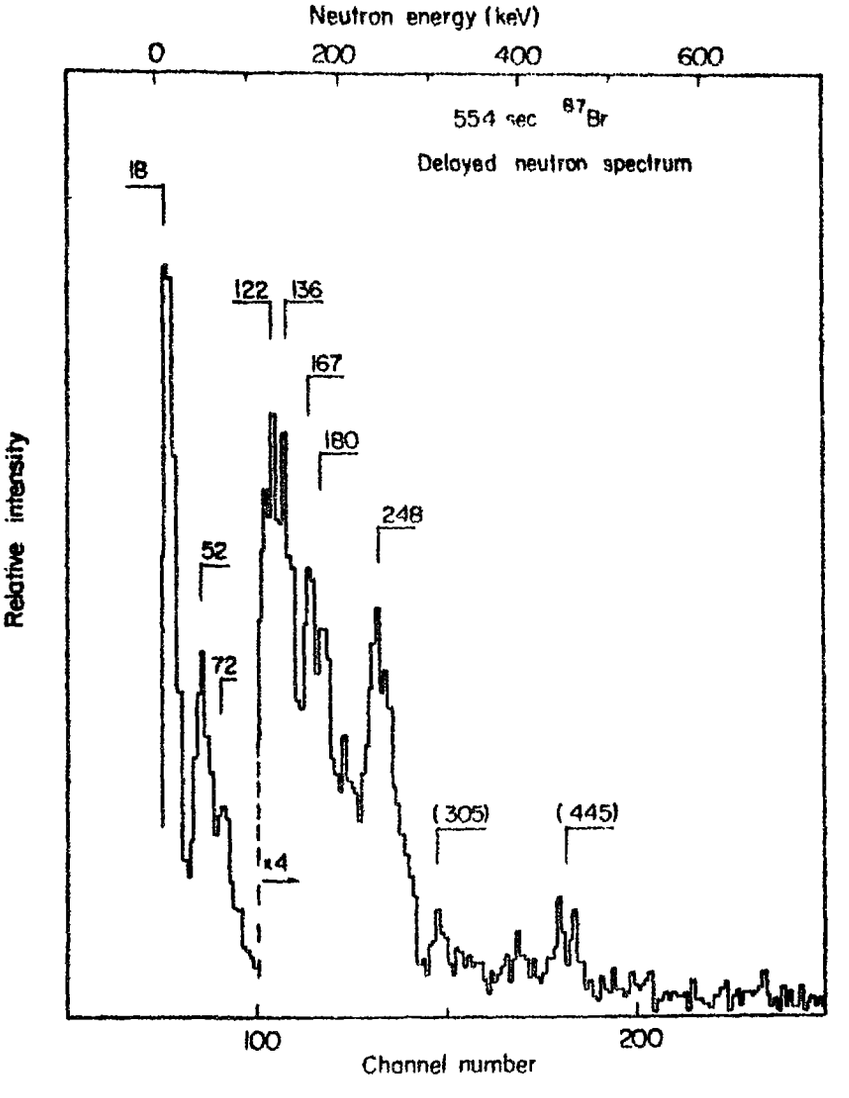

Fig. 5. Delayed neutron spectrum of ^{87}Br obtained by the Mainz group. (Resolution ∿22 keV for energies below 1 MeV, increasing to 45 keV at 2 MeV.)

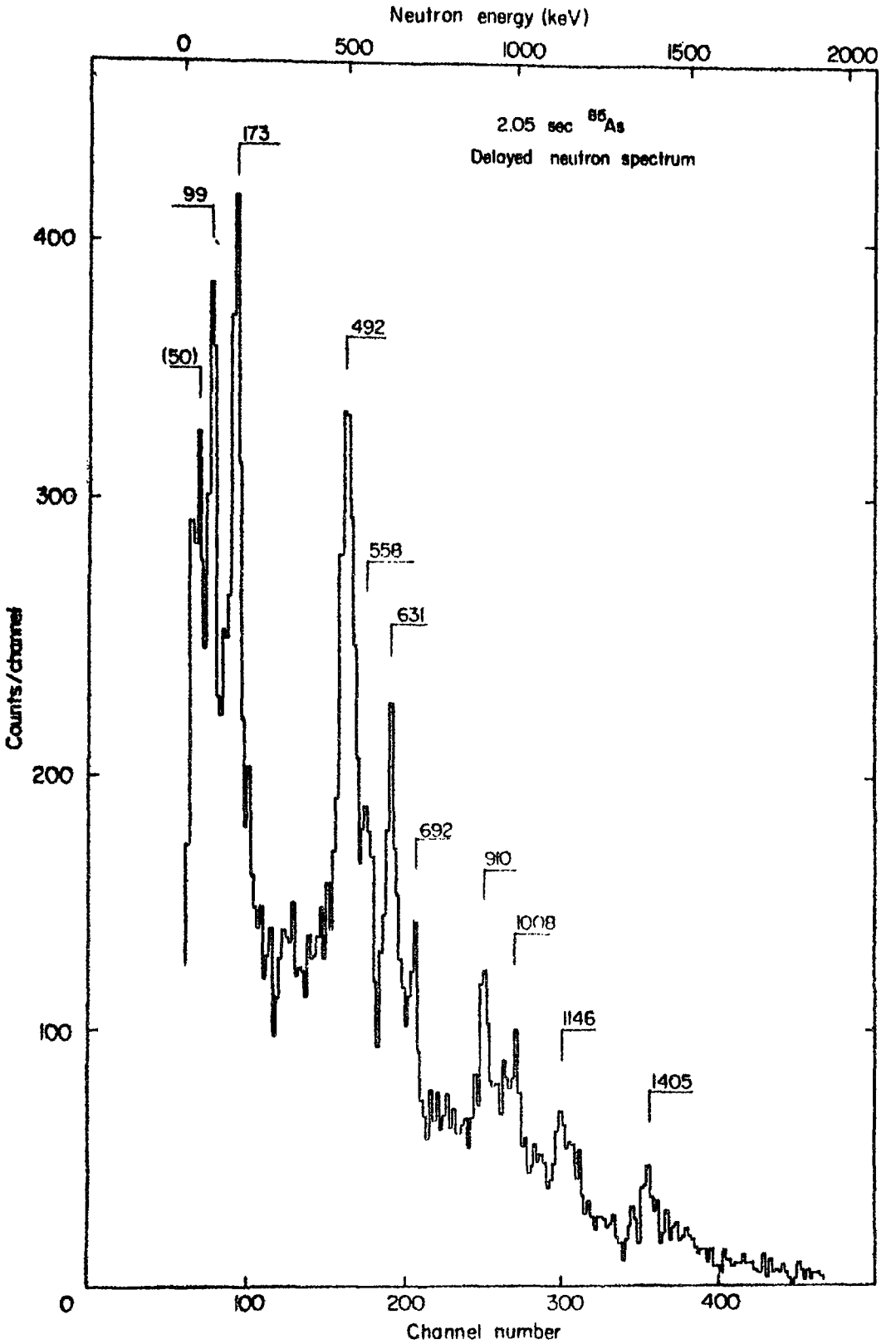

Fig. 6. Delayed neutron spectrum of ^{85}As obtained by the Mainz grou (Resolution ~22 keV for energies below 1 MeV, increasing to 45 ke at 2 MeV.)

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* Contributions published these Panel proceedings, Vol. 3.

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Review Paper no. 14

INTEGRAL DETERMINATION OF NEUTRON ABSORPTION BY FISSION PRODUCTS

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Abstract:

The techniques for integral measurements of fission product capture cross sections are outlined. The presently performed experiments are discussed and the results obtained up to now are critically reviewed.

1. GENERAL ASPECTS

1.1. Introduction

In this paper integral measurements of neutron absorption by fission products are reviewed. The great number of fission product nuclides of importance (over 60), the scarcity of separated materials and the experimental difficulties (radioactivity of target material) preclude for the time being extensive differential measurements.

In the evaluation of fission product absorption cross sections, integrally determined values as reaction rates or reactivity worths in well known neutron spectra, can be used for normalization or adjustment of the evaluated cross sections within the error limits. Moreover, integral quantities may provide empirical data for use in calculations for burnup and fuel management. The present status of the fission product capture cross sections is reviewed in |1|, the present accuracy needs for fast reactor calculations in |2|. In the past Greebler |3| estimated the inaccuracy in fission product absorption in a fast reactor to amount to 40% (90% confidence level). In view of economics this should be reduced to 10%.

It is believed that a combination of the present knowledge of the cross sections (based on resonance parameters and other available differential data and calculations with the best phenomenological nuclear models) with integral measurements might lead to a reduction of the present uncertainties to the required level of 10%.

In an integral cross section measurement the result obtained is an integral of the wanted quantity over the energy weighted with a function depending on the type of measurement.

In an activation or transmutation measurement this weighting function is the neutron energy spectrum $\phi(E)$:

$$\overline{\sigma_{act}} = \frac{\int \sigma_{act}(E)\ \phi(E)dE}{\int \phi(E)dE} \quad . \tag{1}$$

In a reactivity worth measurement this weighting function is the product of neutron spectrum and adjoint: $\phi(E)\ \phi^{+}(E)$:

$$\overline{\sigma_{abs}} = \frac{\int \sigma_{abs}(E)\ \phi(E)\ \phi^{+}(E)dE}{\int \phi(E)\ \phi^{+}(E)dE} \quad . \tag{2}$$

A good knowledge of the weighting function is imperative for using integral data. For limiting the uncertainties in applying the integral data to actual reactor conditions the $\phi(E)$ and $\phi^{+}(E)$ in which the measurements are performed should not differ very much from the same quantities in the reactor system to which the data are applied.

1.2. Classification of experiments considered

Three types of experiments will be considered in this review.

- Reactivity worth measurements.
- Activation measurements.
- Transmutation measurements.

1.2.1. Reactivity worth measurements

Here the difference in the reactivity worth of a test sample and of a standard- or background- sample introduced in the centre of a critical reactor is determined. If the standard is a void the reactivity worth of the sample can, according to first order perturbation theory, be expressed as

$$\delta\rho = \frac{\int_{V_s} \left\{-\int_{E} \Sigma_a(E)\phi(E)\phi^{+}(E)dE - \int_{E}\int_{E'} \Sigma_{sc}(E\to E')\phi(E)\left(\phi^{+}(E)-\phi^{+}(E')\right)dEdE'\right\}d\bar{r}}{\int_{V_r}\int_{E}\int_{E'} \left(\nu\ \Sigma_f(E)\right)\ \phi(E)\ \chi_f(E')\ \phi^{+}(E')dEdE'\ d\bar{r}} \tag{3}$$

in which:

V_s = the sample volume;

V_r = the reactor volume.

The other symbols have their usual meaning.

The assumptions for (3) are

i) The sample is small and located in the centre so that gradient terms can be neglected;

ii) The samples do not contain fissile isotopes. Otherwise a term

$$\int_{V_s}\int_{E}\int_{E'} \left(\nu \Sigma_f(E)\right)_s \phi(E)\, \chi(E')\, \phi^+(E')\,dE\,dE'\,d\bar{r}$$

has to be added to the numerator.

iii) The effects of the sample container and of materials other than in the sample investigated are balanced by a reference sample or otherwise corrected for.

If $\phi(E)$, $\phi^+(E)$ and $\Sigma_{sc\ inelastic}$ are known, the numerator can be corrected for the scattering contribution.

The denominator is the so-called normalization integral. It can be determined by calculations or by measurements with standard materials with well-known cross sections, in which case the cross section to be measured is directly related to the cross section of the standards (see section 1.4.). $\delta\rho$ is deduced from the time behaviour of the neutron flux (inverse kinetic method) or by balancing the reactivity effect of the sample by a special small control member (auto rod technique), which again is calibrated by a kinetic measurement. In all cases where period or inhours are translated into absolute reactivities, the value of the effective β has to be known.

The limit of accuracy in this type of measurements is set by the neutron noise. It has been shown [4] that the relative standard deviation of the detected reactivity is:

$$\frac{\sigma_\rho}{\rho} \sim \frac{10^{-5}}{\sqrt{PT}} \times f$$

in which:

P = reactor power in W;

T = time of measurement in s;

f = form factor to account for the space and energy distributions of the flux and adjoint flux (f is of the order of 1).

In practical cases P is 1 to 10 W and T of the order of an hour, so $\sigma\rho/\rho \sim 10^{-7}$. Higher reactor powers or longer measuring times do not give much improvement due to the increase of drift effects.

For obtaining a reactivity signal well above the background (say 30 to 100 times), samples of fission products of 10 mg to 1 g for thermal systems and 1 g to 100 g for fast systems are needed. The effects of impurities and/or contaminations in the samples can be most disturbing. In fast reactors the reactivity effect of hydrogen can be quite high, so a small contamination with H_2O can lead to erroneous results. In STEK-2000 e.g. an amount of 1 mg of H_2O causes an effect of $.33\times10^{-7}$ $\Delta k/k$ which is equal to the effect of 20 mg of fission products of ^{235}U. In thermal systems a slight contamination with impurities with a large cross section may be disastrous.

The quantity derived from a reactivity worth measurement is of the type

$$\int_{V_s}\int_{E} \Sigma_a(E)\ \phi(E,\bar{r})\ \phi^{+}(E,\bar{r})\,dE\,d\bar{r}. \qquad (4)$$

Due to the neutron absorption in the sample the neutron flux inside and around the sample will be depressed. This is particularly true for neutrons with energies where the cross section of the sample material has resonances. These selfshielding and flux depression are in most cases not negligible due to the finite sizes of the samples. Corrections can be made by performing measurements with different masses of the sample and extrapolation to zero sample mass. However, the smaller the sample the less accurate the reactivity worth is measured, so that for sufficiently small samples mostly no usuable result is obtained. In thermal systems the selfshielding and flux depression can be measured by means of flux mapping using activation foils. For calculating the effect of flux depressions an elegant method has been developed [5].

1.2.2. Activation measurements

If the sample material can be activated by neutron capture the activation cross section (averaged over the neutron spectrum) can be determined from the absolutely measured activity provided the absolute flux is known. The accuracy attainable is limited to that of the absolute activity measurement which may be performed either directly or by comparing with standards. The accuracy obtainable varies from a few per cent in "good" cases to 10% or worse for "bad" cases. "Good" or "bad" refer to intensity and type of radiation to be detected, background radiation from contaminations, etc. The decay scheme of the active nuclide should be well known. Sample amounts in the range of a few to 100 mg are sufficient depending on half-live of the induced activity, the neutron spectrum and the cross section involved. The chemical composition is generally not important.

However, the samples should be free from isotopes which may cause, after the irradiation, a radiation interferring with the radiation of the target sample material. In case the target material itself is already radioactive, a purification just prior to the irradiation might be necessary to remove unwanted daughter products. Since the samples mostly can be small, little trouble with neutron selfshielding and flux depression is to be expected.

Of the about 60 most important fission product nuclides about 20 to 30 can be measured with more or less accuracy by activation in moderate neutron fluxes (10^9-10^{12} $cm^{-2}s^{-1}$).

1.2.3. Transmutation measurements

If sufficient flux is available the resulting shift in isotopic composition due to the transmutation induced by the irradiation can be measured and used in the determination of the cross section. For a moderate flux of 10^{11} cm^{-2} s^{-1} (as in the CFRMF) and for $\bar{\sigma}$ values in the order of ≈ 0.1 to 1 b and for an irradiation time of 1 day $\bar{\sigma}\,\phi\,T \sim 10^{-8}$ to 10^{-9}, so very pure samples have to be used especially in isotopes with masses equal to that of the target nucleus or its capture product (a purity better than 50 ppb is reported to be needed). For irradiations in fast power reactors like Rapsodie, DFR or Phenix, where the fluxes may be 4 orders of magnitudes higher and longer irradiation times up to 100 days are possible, the above mentioned restrictions do not exist. The sample size in all these irradiations may be quite small so no self-shielding will occur. An absolute measurement of the neutron flux (or fluence) is needed in order to deduce a cross section from the experimental result.

1.3. Facilities

Reactivity worth measurements can only be performed in a reactor operated at low power (a few Watt) if drift effects due to temperature variations are to be avoided. If disturbances like temperature changes in the reactor room, movement of cooling water, vibrations, etc. are kept to a minimum, it is possible in reactivity worth measurements to reach the lower limit, set by the neutron noise up to a factor of 2 to 3.

1.3.1. Reactivity measurements in thermal systems

For measurements in thermal spectra, thermal reactors as e.g. the light water moderated swimming pool PTR or the D_2O moderated natural uranium reactor R 0 are used.

The neutron spectrum is mainly Maxwellian. The "temperature" of the Maxwellian can be measured by foil activations. The neutron spectrum can be hardened by heatingthe system as is done for graphite moderated systems (see also the appendix). Deviations of the 1/v behaviour of cross sections can be determined in this way.

The epithermal contribution to the thermal reactor spectrum depends largely on reactor type and on the position of the sample in the reactor. For instance by rearranging the fuel around the sample the epithermal contribution (the Westcott r value) can be changed.
From measurements for two different r values the absorption resonance integral can be deduced. Another way to measure resonance integrals is to perform the measurements in a stationary Cd tube in the reactor where by the thermal contribution to the sample worth is eliminated.
In general, these types of measurements are subject to failure. For a good accuracy of the resonance integral the contribution of the epithermal part to the total signal should be high, i.e. the difference in r values should be high. However, a high r value usually means a deviation of 1/E of the epithermal flux.
When a Cd tube around the sample is used, the determination of the flux and the adjoint flux as function of energy close to the Cd cut-off energy is uncertain. Only if the resonance integral is large compared to the thermal cross section and the lowest resonance lays well above the Cd cut-off may the Cd tube method be used. This also means that the use of boron as standard in this case is subject to the Cd cut-off difficulties.

1.3.2. Reactivity measurements in fast systems

For measurements in a fast spectrum, a fast reactor such as FR 0 or a coupled fast-thermal system as STEK, CFRMF and ERMINE is used.
By varying the composition of the fast core or fast zone the neutron spectrum and adjoint spectrum can be varied. In the coupled fast-thermal systems the sensitivity for a sample introduced in the fast zone is lower than in all-fast systems with the same composition. The sensitivity is reduced at the rate the fast zone contributes to the total reactivity.

1.3.3. Activation measurements

Activation can in principle be performed by any type of neutron source having the desired energy spectrum. In practice, the samples are irradiated in a reactor either directly or with spectrum shaping filters.

1.3.4. Transmutation measurements

Transmutation measurements can only be performed well in systems with sufficient flux ($10^{12} - 10^{15}$ cm^{-2} s^{-1}) e.g. power reactors without or with spectrum filters. An accurate determination of the neutron spectrum under such filters might present a problem. The use of foils of standard materials combined with a spectrum code like SAND-II (see section 1.4.) might offer a solution.

1.4. Normalization problems

For the calibration or normalization of the measurements, standards are used.

For reactivity worth measurements one can for example measure the effects of samples of well-known cross sections. For the activation and transmutation measurements one "only" needs an absolute flux or fluence measurement, to be performed for example by irradiation of one or more detector materials with well-known activation cross sections. In fact one measures

$$\int \sigma_{\text{standard}}\, \phi(E)\phi^{+}(E)dE \text{ or } \int \sigma^{\text{act}}_{\text{standard}}\, \phi(E)dE.$$

Preferably the cross sections of the standard materials should have about the same energy dependency as the material under investigation in order to avoid uncertainties due to an unequal weighting of the flux and the adjoint. In this respect ^{197}Au, which has a rather well established absorption cross section, is not a very suitable standard in fast reactor systems because the important low energy resonances may give relatively too high a weight to the low energy part of the spectrum. For measurements of resonance integrals in 1/E spectra Au is a good standard.

The absorption in fission products in a fast reactor stems partly from capture in the resonance region and partly from capture at higher energies up to several tens of keV's. Therefore a 1/v absorber as ^{10}B, which has a rather well established absorption cross section up to 100-200 keV, could be a suitable standard material [7].
For measurements in thermal systems ^{10}B is a good standard. It can be used in a solution as boric acid or dispersed in Al or C. For the very small quantities, needed in thermal calibrations, it may be diluted with ^{11}B to avoid weighing errors.

For fast systems the use of ^{127}I, the cross section of which is rather well-known, might be considered. Being a fission product itself it has about the required cross section behaviour, furthermore the scattering is not very different from that of the other fission products.

There exist, however, a large uncertainty in the decay scheme of ^{128}I which limits the value of ^{127}I as activation measurement standard.

The fission cross sections of the thermally fissionable isotopes ^{235}U and ^{239}Pu are also rather well established and can be used as standards, albeit that due to the fairly constant or slightly increasing values for σ_f above 100 keV too much weight may be put on the high energy side of the spectrum.

If the fission reaction is not directly used e.g. in an absolute fission chamber but by subsequent γ-counting of some selected specific fission product activity, fission yield uncertainties come into play.

The reactivity effect of fissionable material is mainly composed of two effects, viz. neutron production by fission and neutron losses by absorption, i.e. one measures and uses as standard:

$$+ \int \int \nu\sigma_f(E)\phi(E)\chi_f(E')\phi^+(E')dEdE'' - \int \sigma_a(E)\phi(E)\phi^+(E)dE. \qquad (5)$$

In practice the first term is, in fast systems, 2 to 3 times larger than the second. Since σ_a is less well established, especially for lower energies, an uncertainty is introduced. A variant on this method of standardization is the measurement of the apparent reactivity worth of a calibrated ^{252}Cf neutron source at an absolutely determined fission rate at the position of the source |8|. In fact one measures and uses as standard

$$\rho_o = \frac{\int \sigma_f(E)\phi(E)dE \int \chi_{Cf}(E)\ \phi^+(E)dE}{N.I.}\ , \quad \text{where} \qquad (6)$$

N.I. stands for normalization integral which is the denominator of equation (3).

In equation (6) the flux is weighted only by the $\sigma_f(E)$. The adjoint, however, is weighted by the fission source spectrum χ_{Cf}, which has as a consequence that the adjoint fluxes below 100 keV are no longer taken into account in the weighting. This is just the region where most absorption in fission products in fast systems takes place.

As standard for activation and transmutation measurements the use of ^{238}U is suggested by the CEA Group at Fontenay aux Roses.

Multiple foil activation applied for the measurement of the neutron spectrum can also be used to determine the absolute flux. In this method a great number of foils of activation detector materials for which the cross sections are reasonably known, are irradiated simultaneously. By use of the computer program SAND-II |9| the absolutely determined activations are all used simultaneously to adjust a trial spectrum, thus an adjusted spectrum and an absolute flux level is obtained. Due to the

great number of activation materials, errors in individual cross sections become less important. Moreover, this method can be calibrated in well-known spectra of well-known intensity. This has actually been done for the CFRMF, where, in the framework of the USA Interlaboratory LMFBR Reactor Rate (ILRR) program, the absolute flux has been established.

1.5. Spectrum of neutron flux and adjoint

When the results of the integral measurements are to be used in checking data files or adjusting data files, the neutron spectrum and adjoint spectrum at the location of the sample should be known.
These spectra are calculated by reactor codes with diffusion or transport theories or with Monte Carlo methods. The accuracy obtainable is limited by the accuracy of the calculational models and the cross sections used in the calculation. An experimental determination of the spectrum (and adjoint) may increase the accuracy of the spectrum knowledge.
A variety of methods exists for spectrum measurements ranging from integral measurements of activation- and fission rates, sandwich foil detectors, to differential measurements by time-of-flight techniques, proton recoil technique, ^{6}Li semi-conductor sandwich detectors or ^{3}He proportional counters. For details the reader is referred to |10|.
In general it may be stated that by a combination of each of the methods mentioned an accuracy of the order of 10% over the full energy range seems obtainable.
For the measurements of the adjoint spectrum not many possibilities exist. Korthaus |11| used a number of (α,n) and (γ,n) sources with different averaged neutron energies. However, most sources produce neutrons with energies above 0.4 MeV, only the (Sb-Be) source produces neutrons with substantial lower energy (21 keV).

1.6. Types of samples used

Three types of samples are used.

1.6.1. Separated isotopic samples

In some cases these have been obtained on loan from the US-AEC Research Pool in gram quantities. The isotopic samples, to which also the samples of elements occurring in nature with only one isotope should be reckoned, are used for reactivity worth measurements as well as for activation measurements. The use of these samples is directed at a measurement of the cross section of isotopes for verification of data files

or for adjustment in an evaluation procedure. With these cross sections and known yields |12,13| the absorption of actual fission product mixtures at different burnup stages, can be calculated and a set of pseudo-fission product cross sections derived. Since most samples are not very pure (enrichment 95% or less) reactivity worth measurements are sometimes needed for the other isotopes occurring in the sample in order to be able to correct for their perturbing contribution.

1.6.2. Mixtures of fission product isotopes

These are the samples consisting of bulk fission products prepared by an extensive burnup of fuel samples in power reactors. These samples contain also residual fuel and carrier material since a quantitative separation of fission products from such samples to remove the residual fuel and/or carrier material proved to be not successful |14|.

Mentioned can be here the ^{233}U-Al, ^{235}U-Al, ^{239}Pu-Al Canadian samples, the ^{235}U-Al Dutch (MTR plate) samples, the Pu-C Dutch Dragon coated particles samples. All these samples contain fission products generated by thermal fission of the U or Pu (together with the carrier material). If the amount and isotopic composition of the residual heavy metals are known and corrected for, the effect of the gross fission products can be determined.
To this class of samples belong also samples of actual reactor fuel irradiated to different degrees of burnup in power reactors as SENA, Dragon, Rapsodie, Phenix and EBRII. The reactivity measurements performed or to be performed with these samples in MINERVE (Melodie), HECTOR, MINERVE (Ermine), and CFRMF are more aimed at checking the overall reactivity changes, caused mainly by the changes in the isotopic composition of the heavy metals and only to a lesser extent by fission product formation. Nevertheless useful results for gross fission product absorption may be obtained, provided the quantities of heavy metals are very accurately known. (See appendix).

1.6.3. Mock-up samples of fission products

In the early days of the ZPR, fission products were simulated in material worth measurements by mixtures of natural elements |15|. Later Schröder |16| made substantial improvements composing a mixture simulating the ^{239}Pu fission product of a steam cooled fast reactor. From these mock-up samples no information is obtained that could not be obtained from separate measurements of the various constituents, but these samples have the advantage that due to the rather low abun-

dance of each particular isotope the selfshielding in the total sample is rather low. Thus they can be used as a check on the procedures for calculating cross sections of pseudo fission products and as a check on the analysis of the bulk fission product samples mentioned under 1.6.2.

2. DESCRIPTION OF THE MEASUREMENTS CONSIDERED

2.1. Thermal measurements on gross fission products

2.1.1. PTR

The PTR (Chalk River) [17,18] is a small pool type light water reactor with MTR type fuel. The difference between the reactivity worth of the sample to be measured and that of a reference sample placed alternately in the centre of the core is measured. The samples are placed in a sample carrier which moves in a vertical, water filled, tube. Two sample carriers were used, one of lucite and one of stainless steel thus providing a difference in the neutron spectrum at the location of the samples.

The Westcott r values are 0.029 and 0.047 and the effective neutron temperatures 32°C and 42°C respectively. The fission product samples are alloys of aluminium with ^{233}U, ^{235}U or ^{239}Pu, irradiated in the NRU to about 2.5 n/kb in a thermal spectrum with r = 0.022±0.004 and T_n = (40±5)°C. The samples are right circular cylinders, 1.5 cm diam. and 12 cm high, containing initially about 57 g Al with either 1.5 g ^{233}U, 1.5 g ^{235}U or 1 g ^{239}Pu. After the irradiation, the ^{233}U and ^{235}U samples contained about 0.4 g heavy metals and 0.8 g fission products; for the ^{239}Pu sample these figures were 0.2 g Pu and 0.8 g fission products. Also blanks of the same Al were irradiated and later used in the measurements.

Four samples of each type were irradiated, measurements were only made on two. Extensive destructive analyses were performed both on unirradiated and on the irradiated samples to determine the amount and isotopic composition of the heavy metal before and after irradiation. For the calibration of the oscillator, samples were used made of Al-B, Al-U, Al-Pu plus Al-U-B and Al-Pu-B alloys. The effects of the isotopes not occurring in the calibration samples, i.e. ^{234}, ^{236}, ^{238}U, ^{240}, ^{241}, ^{242}Pu were corrected for by calculations. The estimated accuracy in the measured thermal fission product cross section was 8-10%. In the case

of ^{233}U and ^{235}U the corresponding calculated FISSPROD values, based on input data from |19| and |20|, were higher by more than the compounded errors. Since some systematic error in the PTR pile oscillator measurement was suspected, measurements were repeated in the R 0 (Studsvik) with the two duplicate samples.

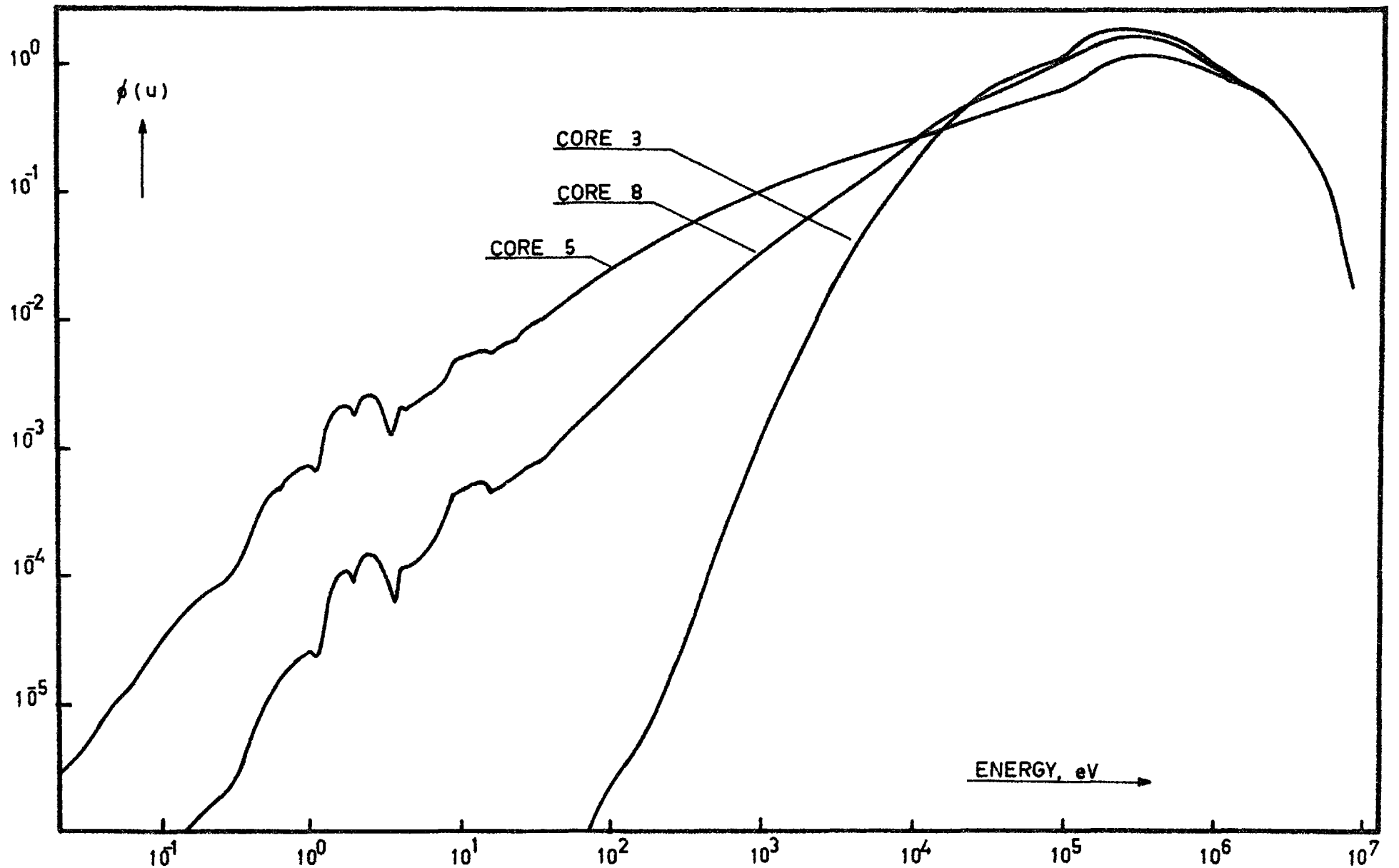
2.1.2. R 0

The R 0 |21| is a D_2O moderated low power reactor with natural uranium metal rods in a square lattice whose geometry can be varied easily. Measurements were made in a thermal column, obtained by removing the 24 innermost rods from a 13 cm square lattice. Measurements were also performed on a cell boundary position.
For the thermal column $T_n = (28\pm3)^oC$, r = 0.00162±0.00005, for the lattice position $T_n = (49\pm3)^oC$ and r = 0.0421±0.0015.
The same calibration samples as in the PTR measurements were used. However, discrepancies were observed in the reactor calibration when using the Al-B samples or samples of boron containing D_2O for the calibration.
In the final report no values for the resonance integrals are given due to unexplained differences between boron and aluminium worth in the high and low r spectra, indicating that the high r spectra data might be unreliable |22|. Results are given in table I and discussed in section 3.1.

Table I: Results of measurements in R 0 of integral fission product capture (σ_o in barns/fission).

Fissile isotope	^{233}U	^{233}U	^{235}U	^{235}U	^{239}Pu	^{239}Pu
Fluence (n/kb)	2.147	2.642	1.888	2.240	2.333	1.915
σ_o (exp.)	41.9±2.5	38.5±1.8	48.7±2.2	46.5±1.6	62.3±3.3	71.1±3.8
σ_o [a] (calc.)	44.2	42.5	52.0	50.3	66.7	68.2
calc./exp.	1.05	1.10	1.07	1.08	1.07	0.96

a) Calculations with latest version of FISSPROD. |23|.



CALCULATED CENTRAL NEUTRON SPECTRA FOR FRO CORES 3, 5 AND 8

FIG.1

2.2. Fast reactor measurements

2.2.1. FRO (Studsvik)

The FRO [24] is a fast critical reactor of the split table type with vertical stainless steel fuel boxes. The fuel consists of 20% enriched uranium platelets. As diluents are used graphite, stainless steel, aluminium and polythene. Measurements were performed in three different core compositions. The neutron spectra of these cores are given in fig. 1. The most central part of the core is built as "pseudo-homogeneous" zone by use of very thin fuel and diluent platelets in order to avoid heterogeneity effects close to the sample.
The pile oscillator moves horizontally through the core centre. The oscillator consists of an aluminium bar (2.4×2.4 cm^2) in an aluminium channel. The bar contains two pockets of $2.15 \times 2.15 \times 11$ cm^3 for the test sample and for the reference sample.
In cores 5 and 8, with comparatively soft neutron spectra, measurements were also made with a copper layer between the oscillator rod and the core to reduce the effect of neutrons being moderated by the sample into the resonance dips of the unperturbed core spectrum. This effect is difficult to treat in perturbation calculations.

The neutron spectra and adjoint spectra were obtained only by calculation. Fine group spectra were produced by the SPENG program in a fundamental mode for a homogeneous medium. With these spectra group cross sections were produced for the different core materials. 25 group spectra for the sample position were calculated by a diffusion code. The energy intervals of the ABBN scheme were chosen (the 26th, the "thermal" group was omitted).

The samples used are listed in table II. Most samples were cylindrical, some were fla.. The sample containers consisted of an aluminium rod with a central hole of a diameter just large enough to hold the available amount of sample material. The samples were dried to remove any traces of H_2O. The reactor power during the oscillations was about 4 W, each sample has been oscillated for 1-2 hours. The reproducibility of the measurements on one day was 2 to 3 10^{-7} $\Delta k/k$ but worse when repeated at longer intervals.

A very thin (0.0065 cm) uranium (93% enriched) platelet has been used as standard for the reactivity worth measurements. In the analysis a correction for the amount of ^{238}U in this sample has been applied.

In order to compare the results of the measurements with calculated results, the reactivity effect of the samples was calculated with

Table II Listing of samples used in FR 0.

material	mass[a] (in g)	enrichment (in atom%)	cyl. sample diameter (in cm)	flat sample thickness (in cm)
^{133}CsF	8.83	100	0.62	
$^{133}Cs_2CO_3$	12.80	100	0.81	
$^{133}Cs_2CO_3$	2.86	100	0.40	
^{99}Tc	1.99	100	0.41	
$^{149}Sm_2O_3$	1.01	97.72	0.25	
$^{147}Pm_2O_3$	2.33	100	0.45	
^{95}Mo	2.27	96.47	0.60	
^{95}Mo	7.69	96.47	1.10	
^{101}Ru	2.41	97.73	0.30	
^{102}Ru	3.90	99.53	0.30	
^{104}Ru	2.41	99.7	0.30	
^{97}Mo	1.90	92.70	0.60	
^{97}Mo	8.00	92.70	1.10	
F.P. mock-up	3.24	-	0.55	
F.P. mock-up	10.02	-	0.95	
^{103}Rh	1.26	100		0.005
^{103}Rh	2.49	100		0.010
^{103}Rh	3.72	100		0.015
^{103}Rh	4.95	100		0.020
^{235}U	2.298	92.99		0.0065

Cylindrical samples: length 10.45 cm. Flat samples: diameter 1.90 cm.
a) Total mass.

a second order perturbation code in 25 energy groups. The group cross sections were collapsed from the fine group cross section set SPENG. The fine group cross sections in SPENG for the resonance region were generated from published resonance data, e.g. |25| and |26|. Above about 1 keV the capture cross sections were obtained mainly from |27|.

The perturbation calculations were performed for spherical samples with radii chosen to give the same mean chord lengths as the actual samples. Perturbed group fluxes were applied for those energy groups (i.e. outside the resonance region) where unshielded sample cross sections were used. The experimental reactivity worths of the samples were corrected by calculated correction factors for the effect of other

materials present in the samples. The sample size correction factor was derived from a comparison of the results for the finite size samples with the results for infinitely small samples (calculated with a first order perturbation program). For some samples calculations were also performed with 34 energy groups. The results differed not more than 2% from the corresponding 25 group calculations. Results can be found in tables IX and X and are discussed in sections 3.2.1.3. and 3.2.2.

2.2.2. CFRMF

The Coupled Fast Reactivity Measurement Facility is a modification of the advanced reactivity measurement facility (ARMF-II), a pool type reactor with MTR type fuel elements, located at the Idaho site of Aerojet Nuclear Company. The modification consists of the insertion of a central fast zone (6" × 6") into the centre of the ARMF. This fast zone consists of a water tight stainless steel can in which a depleted uranium block with a centre hole of about 5cm diameter is placed.
The uranium block is covered with a boral sheet of 1/4". In the central hole there is a filter of highly enriched boron, clad in stainless steel (¼" thickness) and a thin annulus (0.025") of highly enriched uranium, clad in stainless steel. The boral around the ^{238}U block serves as a filter for the thermal neutrons. The ^{238}U filter degrades the incoming fission neutron spectrum by inelastic scattering to a mean energy of about 700 keV. The inner boron layer provides a 1/v absorption near the experimental region, the ^{235}U annulus reduces the energy dependence of the adjoint and thus the influence of the scattering contribution in reactivity worth measurements. Through the central hole runs an empty tube in which the samples to be measured (diameter up to 1½") are inserted.

Two types of measurements are performed, viz. activation and transmutation and reactivity worth measurements.
For the first type of measurement the reactor can be operated at 10 kW power level (integral flux 1.2×10^{11} $cm^{-2}s^{-1}$) with a stability better than 0.5%. For activation measurements a typical sample size is 50 mg of natural composition. For transmutation measurements an isotope separator is available to purify mg quantities to purities of 50 ppb. Samples of over 10 g are needed for reactivity worth measurements because of the low contribution of the fast zone to the total reactivity. The samples are cylinders 14.2 cm high and with diameters of 1.27, 1.90, 2.54 and 3.175 cm.

The reactivity worth is determined from the change in position of the regulating rod, which has been calibrated against period measure-

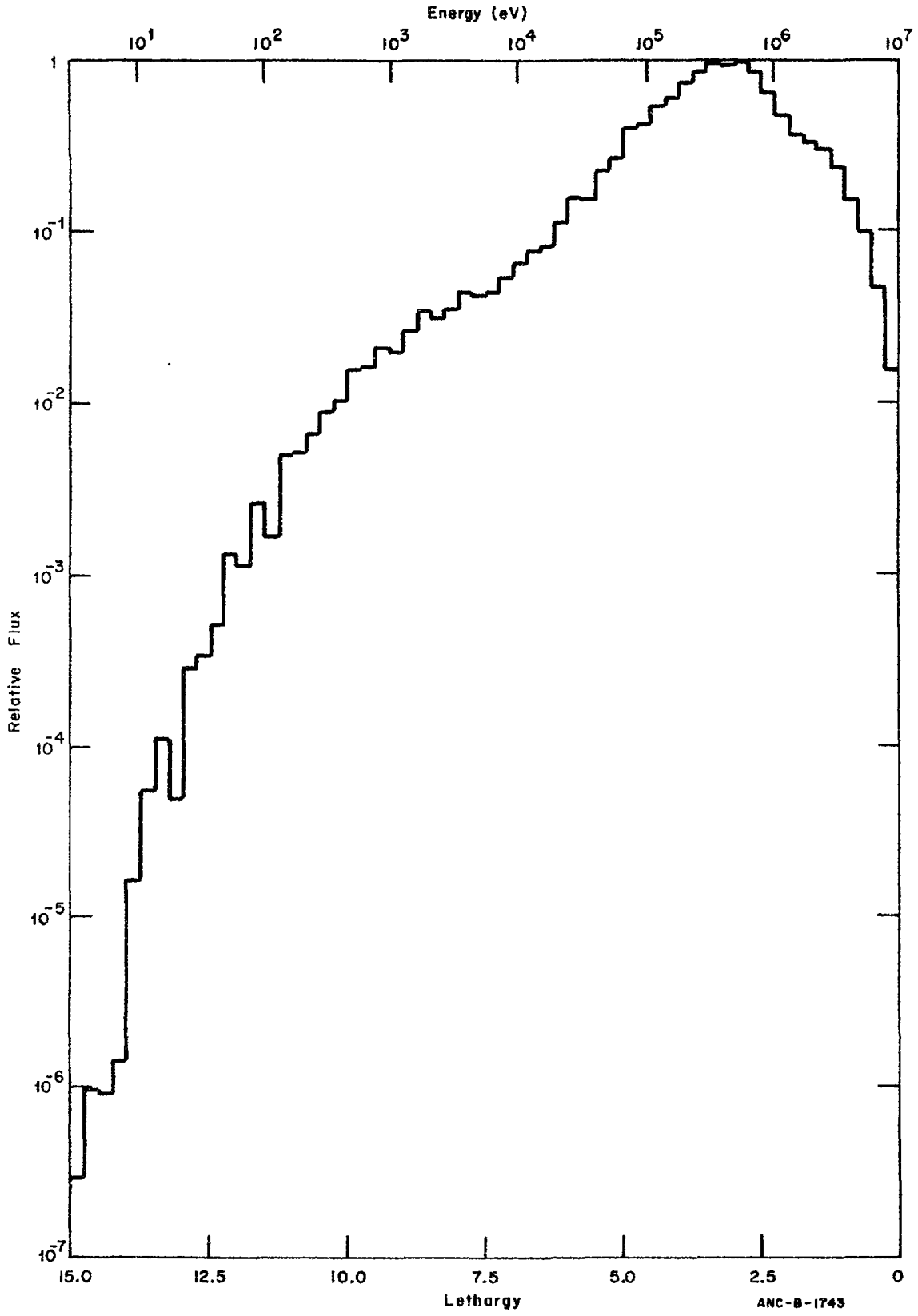


Fig. 2. Calculated neutron spectrum of the CFRMF.

ments. The typical accuracy of these reactivity measurements is ± 2%.

As potential standards are used Au, I, and ^{235}U, which have rather well established cross sections. The low energy sensitivity of Au raises some doubts regarding its suitability as a standard for this type of measurements.

The neutron spectrum at the location of the sample has been measured by proton recoil and multifoil activation.
In the framework of the already mentioned ILRR program the absolute flux at 10 kW has been established with the use of the SAND-II program [9]. The shape of the flux spectrum has also been calculated by 1- and 2-dimensional diffusion programs, 1-dimensional transport and Monte Carlo calculations. The spectrum at the central position is given in fig. 2.

A list of the samples on which measurements have been performed up to now, is given in table III. No transmutation measurements have yet been reported. The results of the measurements are also quoted in tables IIIA and IIIB and are discussed in section 3.2.2.

2.3. STEK

The STEK facility [29,30] was a thermal-fast coupled critical facility located at the Petten site of the RCN (Netherlands). It came into operation in May, 1969 and was closed down on October 3, 1973. STEK was specially built for integral measurements of fission product cross sections by the central reactivity worth method.
In order to have a high reactivity worth per gram of fission product material highly enriched uranium has been used as fuel, while graphite has been used to soften the spectrum to the desired degree. The uranium is in the form of platelets (0.7 mm and 1.4 mm thickness) while the graphite has the form of blocks of about 16 mm thickness. The square cross section of both materials is 50.6×50.6 mm^2.
By stacking graphite and fuel platelets alternately in the thin-walled aluminium fast fuel element boxes, cores with different atomic ratio's of ^{235}U to C are obtained. The C to ^{235}U atomic ratios employed are 11.25 (STEK-500), 22.69 (STEK-1000), 35.14 (STEK-2000), 47.70 (STEK-3000) and 72.48 (STEK-4000).
The fast zone is axially and radially surrounded by lead reflectors around which graphite is stacked. The fast core tank with a diameter of 1 m is surrounded by an annular tank with MTR type fuel elements in water acting as thermal driver. The inner fast zone is always made of such a size that the multiplication factor of the fast zone is

Table IIIA: Listing of fission product isotopic samples measured by activation in CFRMF together with the measured and calculated integral data. [46]

reaction	chemical form [a]	thickness [b] (in mg/cm^2)	$\bar{\sigma}_m$ [c] (barns/target atom)	$\sigma_{calc.}$ [d]
$^{87}Rb(n,\gamma)^{88}Rb$	RbCl	79.7	0.0132 ± 0.0018	
$^{99}Tc(n,\gamma)^{100}Tc$	Tc powder	126.3	0.294 ± 0.039	
$^{102}Ru(n,\gamma)^{103}Ru$	Ru powder	111.4	0.097 ± 0.011	0.125
$^{104}Ru(n,\gamma)^{105}Ru$	Ru powder	96.00	0.0932 ± 0.0093	0.0844
$^{115}In(n,\gamma)^{116}In^m$	In foil	81.11	0.288 ± 0.027	
$^{121}Sb(n,\gamma)^{122}Sb$	Sb powder	168.2	0.309 ± 0.028	0.318
$^{123}Sb(n,\gamma)^{124}Sb$	Sb powder	168.2	0.171 ± 0.024	
$^{127}I(n,\gamma)^{128}I$	HIO_3	249.2	0.304 ± 0.026	0.327
$^{132}Xe(n,\gamma)^{133}Xe^m$	^{132}Xe	0.2868	0.00248 ± 0.00043	0.0441
$^{132}Xe(n,\gamma)^{133}Xe$	^{132}Xe	0.2868	0.0401 ± 0.0040	
$^{134}Xe(n,\gamma)^{135}Xe$	^{134}Xe	0.1060	0.0152 ± 0.0016	0.0069
$^{133}Cs(n,\gamma)^{134}Cs^m$	Cs_2SO_4	88.62	0.0362 ± 0.0069	
$^{133}Cs(n,\gamma)^{134}Cs$	$CsNO_3$	22.94	0.300 ± 0.026	
$^{141}Pr(n,\gamma)^{142}Pr$	Pr powder	101.6	0.080 ± 0.10	
$^{147}Pm(n,\gamma)^{148}Pm$	$^{147}Pm(NO_3)_3$	13.38	0.379 ± 0.059	
$^{147}Pm(n,\gamma)^{148}Pm$	$^{147}Pm(NO_3)_3$	13.38	0.462 ± 0.059	
$^{148}Nd(n,\gamma)^{149}Nd$	Nd_2O_3	77.44	0.118 ± 0.015	
$^{150}Nd(n,\gamma)^{151}Nd$	Nd_2O_3	77.44	0.107 ± 0.024	
$^{152}Sm(n,\gamma)^{153}Sm$	Sm_2O_3	111.3	0.302 ± 0.030	
$^{154}Sm(n,\gamma)^{155}Sm$	Sm_2O_3	111.3	0.121 ± 0.012	

a) Natural abundance elements are used except where the specific isotopes are indicated.

b) Mass includes all constituent elements indicated in chemical form. The real thickness for powdered and compound samples is 0.76 mm and for foils the real thickness can be determined using normal densities.

c) $\bar{\sigma}_m$ is determined by dividing the measured reaction rate $\phi_o\sigma_m$ by ϕ_o = 1.205×10^{11} cm^{-2}s^{-1} (± 8%). ϕ_o is the integral flux as measured using SAND II and the reaction rates measured in the ILRR program. The error assignments are for 67% confidence level and are absolute, i.e. systematic errors have been included.

d) HEDL 8/73 calculation reported in [28].

Table IIIB: Listing of fission product isotope samples measured by the reactivity worth method in CFRMF together with measured integral data. [a]

material	enrichment (in %)	reactivity per gram[b] of four sample diameters (μk)			
		1.27 cm	1.9 cm	2.54 cm	3.175 cm
$^{90}ZrO_2$	97.85	-0.1125	-0.1113	-0.1194	-0.1157
$^{91}ZrO_2$	89.31	-0.2345	-0.2313		
$^{92}ZrO_2$	95.36	-0.2571	-0.2683		
$^{94}ZrO_2$	96.07	-0.2612	-0.2649		
^{95}Mo	96.45	-0.2375	-0.2399	-0.2368	-0.2338
^{96}Mo	96.76	-0.1847	-0.1877	-0.1877	-0.1822
^{97}Mo	94.25	-0.2642	-0.2673	-0.2647	-0.2614
^{98}Mo [c]	98.30			-0.1314	
$^{98}MoO_3$	97.30	-0.3232	-0.3200	-0.3317	-0.3283
^{100}Mo	97.42	-0.1993	-0.1954	-0.1964	-0.1921
$^{147}Sm_2O_3$	98.34	-0.3797	-0.3762		
$^{149}Sm_2O_3$	97.72	-0.6267	-0.6124	-0.5984	
$^{152}Sm_2O_3$	98.29	-0.1613	-0.1599	-0.1593	-0.1559
$^{154}Sm_2O_3$	98.69	-0.1519	-0.1438	-0.1492	-0.1396
^{197}Au		-0.3986	-0.3988	-0.3972	-0.3936
$^{197}Au_2O_3$		-0.410	-0.3856	-0.3857	-0.3850
$^{127}I_2$		-0.2336	-0.2322	-0.2281	-0.2244
$^{235}UO_2$	93.12	+1.008	+0.9952	+0.9930	+0.9839

a) Values are preliminary pending the corrections for oxygen and other impurities.

b) Grams of the metallic component of the sample material.

c) Solid metal sample 2.159 cm diameter x 2.108 cm length.

close to 0.95.

For the reactivity worth measurements the central fuel element is replaced by a special element which can be moved up and down by a driving system. In this element, which is also filled with core material, (i.e. graphite and uranium), a sample can be placed by help of an automatic sample changer. For handling the highly radioactive (up to 10 kCi) mixed fission product samples a shielded lead container is available.

The reactivity worth of the sample is measured by the oscillating tech-

nique against reference samples.

By use of an on-line computer (DDP-516) and an inverse kinetic program the reactivity is calculated as function of time.
Since the moving oscillator element contains fuel a time dependant reactivity signal from this moving fuel is also obtained. To overcome this only the reactivity values obtained 90 s after each transient are used in determining the sample reactivity worths.

Two types of fission product samples have been used in the STEK-program:

- mixed fission products either as mock-up or as mixtures of all fission products from fissioned uranium or plutonium;
- isotopic fission product samples containing one (or a few) isotopes of one element.

Four mixed fission product samples have been used.
The first two (HFR-101 and HFR-102) are samples produced mainly by thermal fission of ^{235}U. They are cut from the MTR type fuel plates of the HFR[1)] at two locations having a burnup of about 60% and 30% FIMA, containing about 100 g aluminium 10.63 or 5.60 g fission products, 3.51 or 9.78 g ^{235}U and 1.88 or 1.02 g ^{236}U. The compositions of these samples were determined by means of chemical analysis of the uranium and 60% of the fission products combined with a detailed burnup calculation.
The third integral sample is produced by thermal fission of ^{239}Pu in Dragon. It has been prepared from a batch of coated particles of PuO_2 and graphite.
The fourth sample is an integral mock-up fission product sample as prepared by Schröder |16|, simulating the fission products of a Pu fueled steam-cooled fast reactor at about 23000 MWd/Te burnup.

In total 57 samples of the second type are available, most of them enriched in a particular isotope. A large part of the separated isotopes (35) has been obtained on loan from the US-AEC Research Pool. Several other samples, five of them being radioactive, were prepared from reactor wastes.
A survey of all samples, except the four mixtures, is given in table IV.
Most of the samples are packed in thin walled stainless steel cylindrical cans closed by welding after the samples have been very thoroughly dried by heating to 800°C. The can height is always 35 mm, the inner diameter varies from 2 to 27 mm depending on the amount of material to be contained.

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1) The HFR is a 45 MW thermal test reactor of the ORR-type located at the Petten site of the RCN.

Table IV: Listing of samples used in the STEK measurements.

material (symbol)	mass (in g)	enrichment (at. percent)	cyl. sample diameter (in mm)	remarks
Zr	1.6390	^{90}Zr - 51.46	40.8×24.7×0.25	platelet 0.25 mm
"	15.5513	" - "	15.1	
ZrO_2	13.7128	" - "	15.06	
$^{90}ZrO_2$	1.9658	" - 97.66	6.1	
"	11.4861	" - "	15.06	
$^{91}ZrO_2$	1.0254	^{91}Zr - 89.31	6.1	
"	1.5411	" - "	7.6	
"	2.8053	" - "	10.06	
$^{92}ZrO_2$	1.6822	^{92}Zr - 96.68	7.6	
"	6.5572	" - "	15.06	
$^{93}ZrO_2$	2.0661	^{93}Zr - 19.53	10.06	
"	3.0644	" - "	15.06	
"	2.9523	" - "	15.06	
$^{96}ZrO_2$	2.3069	^{96}Zr - 58.42	6.1	
"	6.3264	" - "	10.06	
^{93}Nb	15.4230	^{93}Nb - 100	43×43 × 1	platelets 1 mm
Mo	1.8714	natural	42×42×0.1	platelets 0.1 mm
^{92}Mo	0.5880	^{92}Mo - 97.01	4.0	
"	1.3247	" - "	6.1	
^{94}Mo	0.8727	^{94}Mo - 93.93	4.0	
"	1.0912	" - "	4.5	
^{95}Mo	0.6036	^{95}Mo - 96.47	4.0	
"	1.3668	" - "	6.1	
"	3.5421	" - "	10.06	
^{96}Mo	0.4592	^{96}Mo - 96.44	2.0	
"	0.8543	" - "	2.8	
"	1.6493	" - "	4.0	
^{97}Mo	1.2320	^{97}Mo - 94.25	6.1	
"	2.0323	" - "	7.6	
"	3.3182	" - "	10.06	
^{98}Mo	2.2019	^{98}Mo - 98.3	4.5	
"	6.7008	" - "	7.6	
^{100}Mo	0.9585	^{100}Mo - 95.9	4.5	
"	1.8044	" - "	6.1	
"	4.6238	" - "	10.06	
^{99}Tc	0.1822	^{99}Tc - 100	2.0	
"	0.8064	" - "	4.0	
"	1.8934	" - "	6.1	
^{101}Ru	0.6409	^{101}Ru - 97.73	2.0	
"	2.3230	" - "	4.0	
^{102}Ru	0.6979	^{102}Ru - 96.89	2.8	
"	1.6137	" - "	4.0	
^{104}Ru	0.5926	^{104}Ru - 99.7	2.0	
"	1.9765	" - "	4.0	
^{103}Rh-Al	2.4774	^{103}Rh - 100	43×42.5×0.5	platelets 0.5 mm 4 w/o Rh
^{103}Rh	1.1467	^{103}Rh - 100	43×43×0.05	platelets 0.05 mm and 0.1 mm
^{103}Rh		^{103}Rh - 100	0.5	wire, 0.0244 g/cm

Table IV (continued):

material (symbol)	mass (in g)	enrichment (at. percent)	cyl. sample diameter (in mm)	remarks
Pd		natural	0.5	wire, 0.02388 g/cm
^{104}Pd	0.2765	^{104}Pd - 89.75	2.8	
"	0.2953	" - "	2.8	
^{105}Pd	0.9208	^{105}Pd - 94.51	4.5	
"	1.2964	" - "	6.1	
^{106}Pd	1.3502	^{106}Pd - 96.66	4.0	
^{107}Pd	1.6316	^{107}Pd - 15.70	4.5	
"	2.8954	" - "	6.1	
"	4.7670	" - "	7.6	
^{108}Pd	0.6762	^{108}Pd - 98.11	4.0	
"	1.0064	" - "	4.5	
^{110}Pd	0.2884	^{110}Pd - 96.98	2.8	
"	0.2961	" - "	2.8	
^{109}Ag	0.3285	^{109}Ag - 99.26	2.0	
"	1.1305	" - "	4.0	
^{111}CdO	0.1950	^{111}Cd - 95.29	2.0	
"	0.2014	" - "	2.0	
"	0.3976	" - "	2.8	
In	13.1166	natural	42.5×42.5×1	platelets 1.0 mm
^{128}Te	4.9361	^{128}Te - 99.46	7.6	
^{130}Te	1.8713	^{130}Te - 99.49	7.6	
"	3.0308	" - "	10.06	
$Pb^{127}I_2$	0.4336	^{127}I - 100	2.80	
"	1.0209	" - "	4.50	
"	3.1277	" - "	7.60	
$Pb^{129}I_2$	0.9030	^{129}I - 86.1	4.5	
"	1.7974	" - "	6.1	
^{131}Xe	2.970	^{131}Xe - 41.31	sphere, 42	
$^{133}CsCl$	0.9918	^{133}Cs - 100	4.0	
"	2.2296	" - "	6.1	
"	6.0016	" - "	10.06	
"	12.9431	" - "	15.06	
$^{133}CsNo_3$	5.4638	^{133}Cs - 100	10.06	
$^{137}CsCl$	0.9674	^{137}Cs - 33.77	4.5	
"	1.8085	" - "	6.1	
"	3.4285	" - "	7.6	
"	5.8041	" - "	10.06	
$^{139}La_2O_3$	5.4665	^{139}La - 99.911	10.06	
"	7.7975	" - "	15.06	
"	24.6470	" - "	27.0	
$^{140}CeO_2$	13.7840	natural	15.06	^{140}Ce 88.88 a/o
"	38.7447	" - "	27.00	
$^{142}CeO_2$	3.8585	^{142}Ce - 92.11	7.6	
"	6.9082	" - "	10.06	
$^{141}Pr_6O_{11}$	4.9766	^{141}Pr - 100	10.06	
"	9.6044	" - "	15.06	
"	9.4261	" - "	15.06	
"	25.8065	" - "	27.0	

Table IV (continued):

material (symbol)	mass (in g)	enrichment (at. percent)	cyl. sample diameter (in mm)	remarks
Nd	0.6104	natural	4.0	
"	3.1654	"	10.06	
Nd_2O_3	3.236	natural	10.06	
"	3.7364	"	10.06	
"	5.865	"	15.06	
"	22.8048	"	27.0	
$^{142}Nd_2O_3$	1.798	^{142}Nd - 96.21	7.6	
"	1.9767	" - "	10.06	
"	4.3009	" - "	15.06	
$^{143}Nd_2O_3$	0.6724	^{143}Nd - 91.06	4.0	
"	4.6364	" - "	10.06	
$^{144}Nd_2O_3$	1.4722	^{144}Nd - 94.5	2.6	
"	2.5295	" - "	10.06	
"	5.8087	" - "	15.06	
$^{145}Nd_2O_3$	0.3685	^{145}Nd - 91.82	2.8	
"	1.6272	" - "	6.1	
"	2.5292	" - "	7.6	
$^{146}Nd_2O_3$	3.6196	^{146}Nd - 94.5	6.1	
"	7.4224	" - "	10.06	
$^{148}Nd_2O_3$	0.7152	^{148}Nd - 87.9	4.5	
"	1.4268	" - "	6.1	
"	3.9100	" - "	10.06	
$^{150}Nd_2O_3$	0.4064	^{150}Nd - 92.5	2.8	
"	1.2100	" - "	4.5	
"	2.0968	" - "	6.1	
"	3.2441	" - "	7.6	
$^{147}Pm_2O_3$	0.6215	^{147}Pm - 88.07	4.5	On 1-1-1972; $t_{\frac{1}{2}}$=2.62 a
"	1.1216	" - "	6.1	
"	1.8521	" - "	7.6	
$^{147}Sm_2O_3$	0.5098	^{147}Sm - 100	4.0	
"	0.6988	" - "	4.5	
"	1.1960	" - "	6.1	
$^{147}Sm_2O_3$	0.2484	^{147}Sm - 98.34	2.8	
"	1.1756	" - "	4.5	
"	3.1680	" - "	7.6	
$^{148}Sm_2O_3$	0.5100	^{148}Sm - 96.06	2.8	
"	1.0066	" - "	4.0	
"	1.2460	" - "	4.5	
$^{149}Sm_2O_3$	0.3015	^{149}Sm - 97.6	2.0	
"	0.5786	" - "	2.8	
"	1.0206	" - "	4.0	
"	2.5424	" - "	6.1	
$^{150}Sm_2O_3$	0.2248	^{150}Sm - 94.74	2.8	
"	0.4887	" - "	4.0	
"	0.5292	" - "	4.5	
"	1.0570	" - "	6.1	
"	1.6340	" - "	7.6	

Table IV (continued):

material (symbol)	mass (in g)	enrichment (at. percent)	cyl. sample diameter (in mm)	remarks
$^{151}Sm_2O_3$	0.3898	^{151}Sm - 6.13	2.8	
"	0.8670	" - "	4.5	
"	2.6180	" - "	7.6	
$^{152}Sm_2O_3$	0.2366	^{152}Sm - 98.29	2.0	
"	0.4730	" - "	2.8	
"	1.2169	" - "	4.5	
"	2.2912	" - "	6.1	
$^{154}Sm_2O_3$	0.9843	^{154}Sm - 98.69	4.0	
"	1.1206	" - "	4.5	
$^{151}Eu_2O_3$	0.5456	natural	4.0	^{151}Eu - 47.82
"	1.2474	"	6.1	
"	3.0100	"	10.06	
$^{153}Eu_2O_3$	0.4276	^{153}Eu - 98.76	4.0	
"	1.0914	" - "	6.1	
$^{156}Gd_2O_3$	0.2255	^{156}Gd - 99.82	2.0	
"	0.2146	" - "	2.0	
$^{157}Gd_2O_3$	0.2531	^{157}Gd - 93.7	2.0	
"	0.2289	" - "	2.0	
^{159}Tb	0.2782	^{159}Tb - 100	43×43×0.025	foils, 0.025 mm
^{235}U	4.4872	^{235}U - 89.43	43×43×0.14	plates STEK fuel: 0.14 mm
^{235}U	23.4483	^{235}U - 89.93	43×43×0.7	plates 0.7 mm
^{235}U-Al	3.0087	^{235}U - 89.78	41×41×0.6	plates U-Al alloy; U: 11.1 w/o
$^{236}U_3O_8$	1.7546	^{236}U - 32.48	34.4	height 0.5 mm
"	1.7543	" - "	34.4	"
"	1.7546	" - "	34.4	"
"	1.7545	" - "	34.4	"
^{238}U-Al	0.5825	^{238}U - 99.28	43×43×0.1	plates U-Al alloy U: 23.0 w/o
^{238}U	4.8391	^{238}U - 99.59	43×43×0.14	plates 0.14 mm
$^{239}PuO_2$	0.459	^{239}Pu - 98.34	7.5	PuO_2 in PbO
"	1.388	" - "	7.5	"
"	1.952	" - "	7.5	"
"	2.755	" - "	7.5	"
$^{240}PuO_2$	0.082	^{240}Pu - 98.30	7.5	"
$^{240}PuO_2$	0.175	^{240}Pu - 45.70	7.5	From Dragon in PbO
"	0.453	" - "	7.5	" "
"	0.906	" - "	7.5	" "

The mixed fission product samples made out of HFR plates are contained in a stainless steel box $4.3 \times 4.3 \times 4.3$ cm^3. As this box could not be closed by welding it was soldered. Some doubt as to a possible water contamination still exists. A final destructive analysis of the HFR samples will be made at the end of the experiments.

The central spectra in STEK were calculated by a two dimensional diffusion code in 26 groups (ABBN scheme). Several spectral indices were measured by foil activation and use of a double fission chamber. Differ-

ential spectrum measurements have been performed by the triple foil technique, by a time-of-flight technique over a 30 m and a 50 m flight path, by proton recoil counters, by a ^{3}He proportional counter and by ^{6}Li solid state sandwich detectors.

The calculated spectra in four STEK cores together with measurements are shown in fig. 3. In the preliminary analysis of the STEK reactivity worth measurements the calculated STEK spectra were used except for the STEK-4000 core, where below 50 eV the spectrum as measured by the time-of-flight method was used.

The adjoint spectrum has been measured in one core (STEK-3000) with several (α,n) sources and one (γ,n) source. The result is given in fig. 4.

The reactivity worth measurements are normalized by the ^{252}Cf source method (see section 1.4.). The resulting quantity is thus

$$\left(\frac{\rho}{\rho_o}\right)_{capt} = \frac{\int \sigma_c(E)\ \phi(E)\ \phi^{+}(E)dE}{\int \sigma_f(E)\ \phi(E)dE \int \chi_{Cf}(E)\ \phi^{+}(E)dE}\ , \qquad (7)$$

in which

σ_c = microscopic capture cross section of sample isotope, for mixed fission product samples: fission product capture cross section per fissioned atom;

σ_f = microscopic fission cross section of ^{235}U;

χ_{Cf} = ^{252}Cf spontaneous fission neutron spectrum, $\int \chi_{Cf}(E)dE = 1$;

ϕ,ϕ^{+} = flux and adjoint flux at the position of the sample.

The experimental data have to be corrected for a number of effects, i.e.

- reactivity contributions from packing materials and from other isotopes in the sample. This correction is derived from separate measurements with reference samples or from calculations;
- neutron down scattering by the sample. The correction for this effect is made by calculations using the Australian cross section set |31|;
- selfshielding and flux depression.
  To correct for this effect measurements have been made with different sample sizes in order to extrapolate to zero mass. This procedure proved to be unreliable, especially for the measurements in soft neutron spectra where the selfshielding is high. The correction will now be derived from calculations, e.g. by the code TRIX. For the flux depression calculations a modified version of the 1-dimensional transport code DTF-IV is used, which calculates the flux depression $\Delta\phi$ directly.

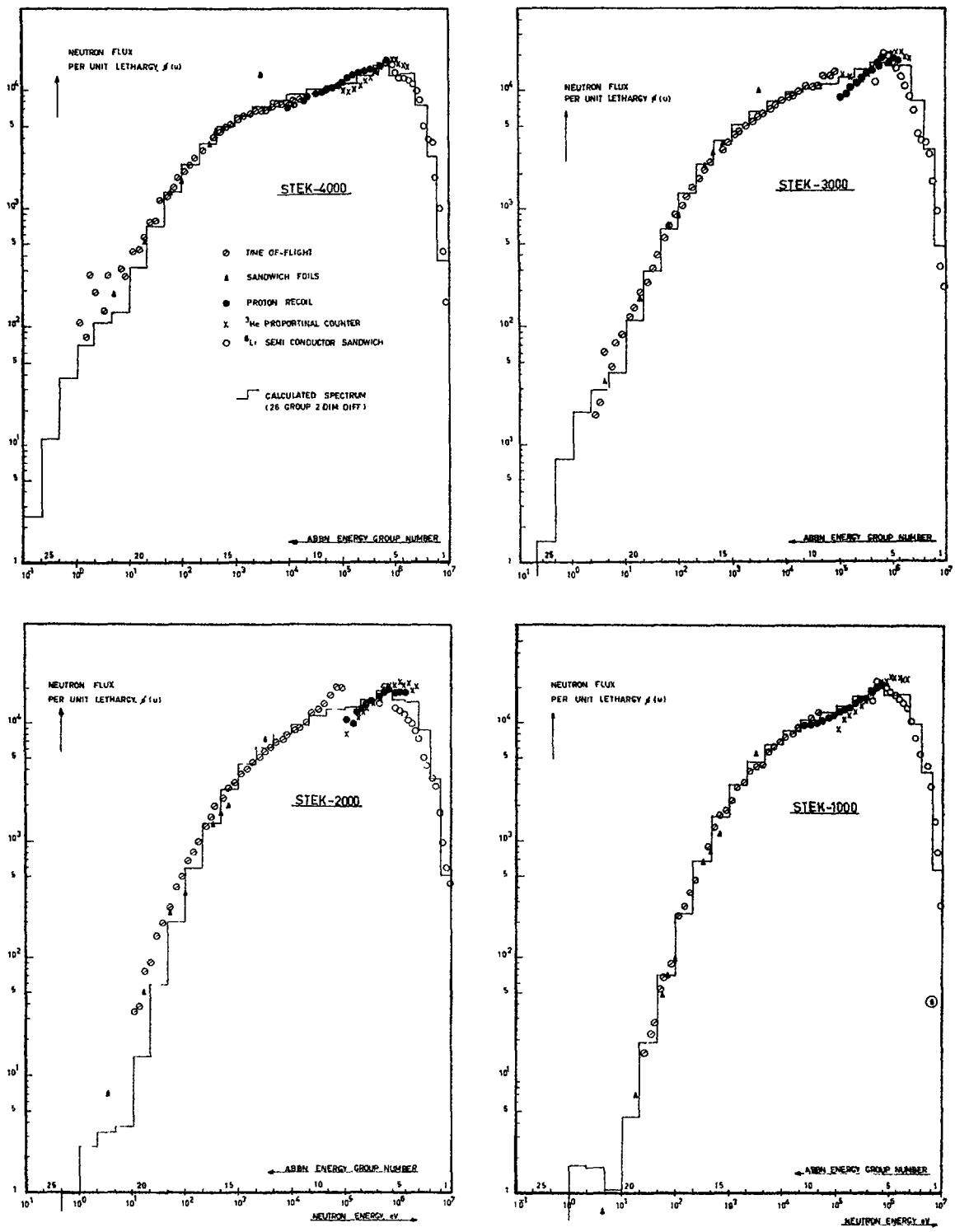


Fig. 3. STEK NEUTRON SPECTRA

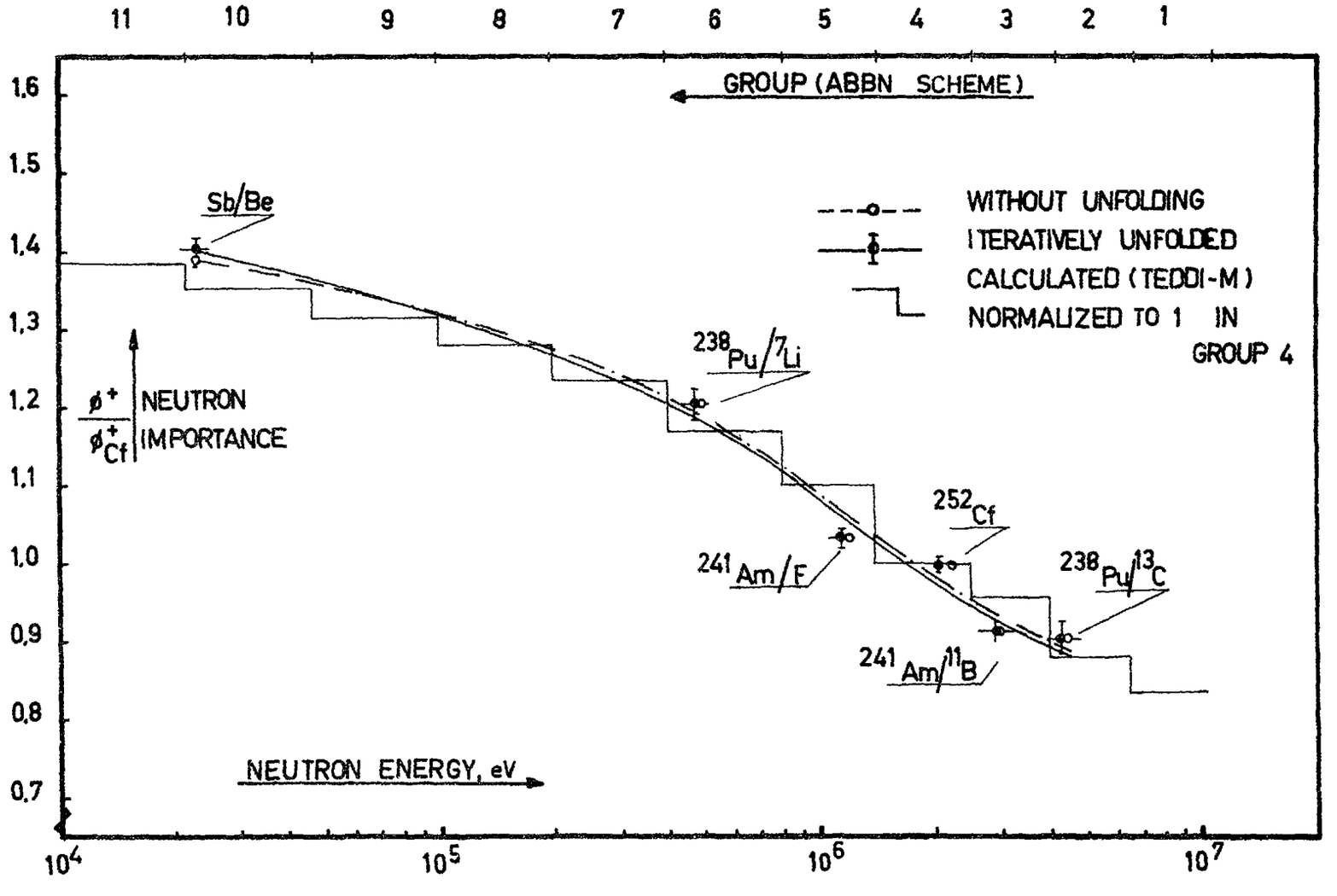


fig. 4 NEUTRON IMPORTANCE IN CORE STEK-3000

Only the measurements with three of the four mixed fission product samples have been analyzed in detail. The results of the Dragon sample could not be used because of uncertainties in composition and in the correction for the reactivity worth of the plutonium (including the selfshielding thereof) in the sample. No results of the very recent measurements in the STEK-500 core can be given. The results for the mixed fission product samples are given in table V and are discussed in detail in section 3.2.1.1.
Some preliminary results on the isotopic samples are contained in table X and are discussed in section 3.2.2.

3. DISCUSSIONS OF THE EXPERIMENTAL RESULTS PRESENTED IN CHAPTER 2

3.1. Measurements on gross fission products in thermal spectra

There appears to be only one good measurement, i.e. that in R 0. There is besides the mentioned PTR measurement also a measurement, reported in |32|, on the reactivity worth of irradiated fuel. This measurement is not taken into account in this review because in that measurement the concentrations of the remaining heavy nuclides have not been measured but were infered from burn-up calculations. However, the main error source in this type of measurements is in fact the uncertainty in the concentration of the remaining fissile material. The effective cross section of this residual fuel, $\nu\sigma_f W - \sigma_a$ (in which W is the importance of fission neutrons relative to thermal neutrons), is an order of magnitude larger than the averaged cross section of fission products, so any error in the amount of remaining fissile material is greatly amplified.

As is seen from table I the values calculated from the FISSPROD library are about 7% higher than the experimental ones. An exception forms the low burn-up ^{239}Pu sample. For that sample, however, the concentrations of the remaining heavy nuclides were obtained by a considerable extrapolation from earlier chemical analyses on equivalent samples.

To improve the accuracy of this type of measurements the effect of the uncertainty in the amount of residual fissile material should be reduced. The present limit for the accuracy of the chemical analyses of the irradiated sample as well as the calibration sample is in the order of tenths of a percent. A considerable improvement in these figures seems not likely to be achieved. An inaccuracy for the residual fissile material of the order of 0.5% or slightly better leads to an

Table V: Measured and calculated reactivity worths of integral samples in STEK and their errors.

1 core	2 sample	3 measured ρ/ρ_o	4 correction for self-shielding and flux depression	5 ρ/ρ_o extrapolated to zero mass	6 ρ/ρ_o of 0+N	7 $(\rho/\rho_o)_{scatt.}$	8 experimental $(\rho/\rho_o)_{capt.}$	9 calculated $(\rho/\rho_o)_{capt.}$	10 calc./exp.	11 error I (in %)	12 error II (in %)	13 error III (in %)	14 error IV (in %)	15 error V (in %)
4000	HFR-101	-.456±.007	1.043±.025	-.475±.014	-	+.020±.006	-.495	-.465	0.94	2.7	3.5	8.2	2.5	4.4
	HFR-102	-.501±.017	1.078±.031	-.540±.024	-	+.020±.006	-.560	-.492	0.88	4.2	4.3	8.2	2.5	4.3
	KFK-1	-.514±.005	1.191±.026	-.603±.018	+.0131±.0005	+.015±.005	-.631	-.679	1.08	2.8	0	8.2	2.3	5.3
	KFK-2	-.483±.003	1.245±.031											
3000	HFR-101	-.353±.005	1.038±.025	-.367±.011	-	+.034±.010	-.401	-.360	0.90	2.5	4.5	7.2	3.3	5.2
	HFR-102	-.397±.025	1.050±.032	-.417±.029	-	+.034±.010	-.451	-.374	0.83	6.3	5.5	7.2	3.1	5.1
	KFK-1	-.356±.006	1.089±.010	-.388±.004	+.0200±.0003	+.026±.008	-.434	-.460	1.06	1.0	0	7.2	2.7	6.0
	KFK-2	-.349±.003	1.115±.010											
2000	HFR-101	-.298±.005	1.024±.025	-.302±.009	-	+.040±.012	-.341	-.302	0.88	2.5	5.6	6.1	4.1	6.5
	HFR-102	-.356±.024	1.032±.031	-.367±.027	-	+.040±.012	-.407	-.311	0.76	6.4	6.8	6.1	3.6	6.5
	KFK-1	-.285±.006	1.044±.015	-.293±.006	+.0272±.0003	+.031±.009	-.350	-.364	1.04	1.6	0	6.1	3.2	7.7
	KFK-2	-.275±.004	1.056±.015											
1000	HFR-101	-.219±.005	1.021±.029	-.223±.008	-	+.057±.017	-.280	-.254	0.91	2.9	7.7	5.2	6.2	7.7
	HFR-102	-.260±.033	1.019±.032	-.254±.035	-	+.057±.017	-.321	-.261	0.81	10.3	10.3	5.2	5.5	7.6
	KFK-1	-.174±.007	1.032±.010	-.185±.004	+.0387±.0004	+.043±.013	-.267	-.304	1.14	1.4	0	5.2	4.8	9.3
	KFK-2	-.179±.004	1.039±.010											

Note: The definition of ρ/ρ_o is according to section 2.3, i.e. for fission product cross sections in barns/fission and σ_f (^{235}U) in barn.

uncertainty in the fission product cross section in the order of 5% or slightly better as a lower limit. An improvement might be achieved by performing the measurements in such a system that the W value is low. The W value for the PTR measurement in the stainless steel sample holder was of the order of 1.6, in the lucite holder of the order of 1.3, while in the R 0 experiment the W values for the thermal column and the lattice configuration were about 1.0 and 0.8 respectively. A W value slightly below 0.5 would be needed for a cancellation of the effect of residual fuel.

3.2. Measurements in fast spectra

3.2.1. The results of measurements on integral samples

3.2.1.1. STEK measurements and their analysis. The measured reactivity worths, obtained with the three mixtures of fission product isotopes, are compared with calculated values based on the RCN-1 capture cross section set evaluated by RCN |33|. All results have been combined in an adjustment calculation taking into account errors and correlations both in the cross sections and in the measurements. These results represent adjusted pseudo fission product cross sections for four mixtures simulating the fission products of ^{235}U, ^{238}U, ^{239}Pu and ^{241}Pu in the SNR-300 reactor.

The experimental results for the three samples are summarized in Table V. The HFR samples contain some moisture, the amount of which is not accurately known at present. Experimental results are somewhat uncertain and provisional, therefore, and will be improved in future.
The selfshielding correction has been calculated for the fission product isotopes with the code TRIX, for the other isotopes in the samples and reference sample the ABBN selfshielding factors have been used. The flux perturbation has been calculated with DTF-IV for two STEK cores. Corrections not calculated directly have been obtained by interpolation or extrapolation or reasonable guessing.
The scattering correction was calculated with data from the Australian set |31|. Five different relative errors (standard deviations) are given in table V. Error I contains the experimental error (column 3), the extrapolation error (column 4) and the error in ρ/ρ_o of oxygen and nitrogen present in the KFK-mixture (column 6). These errors are uncorrelated for different measurements. Error II contains all errors due to uncertainties in sample composition (uranium and fission product contents, isotopic composition and moisture). These errors are fully

correlated for measurements with the same sample in different cores, but are uncorrelated for different samples. Errors III are fully correlated for measurements in the same core, and include the uncertainty in ρ/ρ_o due to spectrum uncertainties and errors in the measured normalizations (ρ_o). Error IV is fully correlated for all measurements with all samples in all cores, due to common errors in ρ_o (^{252}Cf source strength, error in σ_f of ^{235}U) and to the uncertainty in the scattering correction, which was assumed to amount to 30%.

Finally, error V is the relative error due to cross section uncertainties, to be discussed in section 3.2.1.4. From errors I to IV a complete covariance matrix of the 12 corrected experimental values has been calculated.

The RCN-1 evaluation |33| of group cross sections of 75 fission products in ABBN format |34| was used to calculate the group constants of fission product mixtures. The Australian set of Rose |31| was used to supplement the RCN-1 set for some of the isotopes.
Also the inelastic scattering matrices are entirely based on |31|.

Shortcomings of the RCN-1 model arise typically above 0.2 MeV. However, for a mixture of fission products, the group cross sections of RCN-1 are only systematically too high for energies above 1 to 2 MeV. Therefore, these model errors are of minor importance for the interpretation of mixed fission product sample reactivity measurements and for the calculation of integral fission product quantities in a fast reactor. For the interpretation of isotopic sample measurements, on the other hand, these model errors will have to be reduced. At any rate, systematic model errors were included in the calculations. One of the advantages of the RCN-1 set is that a rather complete error calculation has been performed for this set |35| for use in an adjustment procedure (see section 3.2.1.4.).

For the three mixtures HFR-101, HFR-102, KFK mock-up, capture group cross sections were calculated with the RCN-1 set using a weighting appropriate to the STEK experiments namely a smoothed STEK-2000 spectrum, The measured and calculated compositions for the HFR samples were used while the composition of the KFK sample was given by the GfK.
For 15 isotopes occurring in the KFK sample for which no cross section data were available in the sets |31,33,36|, an estimate was made about their contribution to the reactivity effect. At a later date an ad hoc evaluation of these cross sections was made which led to some improvements. These improvements have been taken into account in table VI (indicated by RCN*-1).
Also other sets were used to compare measured and calculated reactivities

Table VI: Comparison of experimental capture reactivity worths of integral samples in STEK with results calculated for different sets[a].

The worths are expressed in units of $-\rho/\rho_o$, as defined in section 2.3.

sample	set	core STEK-4000	core STEK-3000	core STEK-2000	core STEK-1000
HFR-101	Exp.	0.495	0.401	0.341	0.280
	RCN-1	0.465±4%	0.360±5%	0.302±7%	0.254±8%
	RCN-1 adj.	0.473±3%	0.365±4%	0.306±5%	0.257±6%
	Benzi/RCN	0.463	0.359	0.301	0.252
	UK NDL	0.555	0.427	0.346	0.281
	Australian	0.424	0.337	0.287	0.246
HFR-102	Exp.	0.560	0.451	0.407	0.321
	RCN-1	0.493±4%	0.374±5%	0.311±7%	0.261±8%
	RCN-1 adj.	0.503±4%	0.381±4%	0.316±6%	0.264±6%
	Benzi/RCN	0.494	0.376	0.313	0.262
	UK NDL	0.575	0.438	0.353	0.286
	Australian	0.453	0.354	0.298	0.255
KFK	Exp.	0.631	0.434	0.350	0.267
	RCN-1	0.676±5%	0.458±6%	0.364±7%	0.306±9%
	RCN-1 adj.	0.629±4%	0.423±6%	0.333±5%	0.277±5%
	RCN-1*	0.633	0.438	0.353	0.298
	Benzi/RCN*	0.634	0.438	0.353	0.298
	UKNDL/RCN*	0.705	0.513	0.417	0.340
	Australian/RCN*	0.545	0.389	0.322	0.274

a) The sets RCN-1, UKNDL and Benzi |33,36,42| have been supplemented, if necessary, with cross sections from the most complete set, which is the Australian set |31|. At energies below 1 keV Benzi's set has been extended with the RCN-1 cross sections. Recently, the RCN-1 set has been updated to include 15 isotopes which are only present in the KFK-sample. This set is indicated with an asterisk.

i.e. the UKNDL set |36|, the Australian set |31|, the set of Benzi |37| for groups 1 to 14, which for the groups 15-26 has been supplemented by data taken from the RCN-1 set. This comparison is given in table VI. From this table it can be concluded that the measured reactivity worths of the HFR samples are systematically higher than most calculated values. Only in the case of the UKNDL set are the calculated values higher than those measured for the cases, HFR-101 in STEK-4000, -3000, -2000, -1000 and HFR-102 in STEK-4000.
The KFK sample is predicted on the whole rather well by the RCN-1* set. The UKNDL set again leads to an overprediction, the Australian set is in most cases to an underprediction.

The group cross sections for the HFR-101 sample as calculated with the different group sets are compared in fig. 5.

3.2.1.2. Pseudo-fission products. For many reactor applications it suffices to combine the fission products to one single fictitious material, which has the same group cross sections as the sum of the actual fission products, the so-called pseudo fission product.

For thermal reactors usually more than one pseudo fission product is needed to describe the formation and the burn-up of fission products during long irradiation times. The burn-up is significant due to the high thermal capture cross section some fission products exhibit.
Since the cross section of fission products in fast reactors are much smaller, for most applications it will suffice to use only one pseudo fission product per fissile isotope.

To obtain the isotopic compositions of the fission products of ^{235}U, ^{238}U, ^{239}Pu and ^{241}Pu after an irradiation of 50 MWd/kg metal, needed for composing the pseudo fission products, zero-dimensional burn-up calculations were performed |45| starting from the average initial material composition of SNR-300 and using the average neutron flux spectrum |38|. The fission product chains taken into account contained 160 isotopes; fission yields and decay branching ratios of |39| and neutron capture branching ratios from |40| were used. The resulting compositions showed indeed only a small dependence on irradiation time.

In table VII the calculated RCN-1 capture group cross sections of the four pseudo fission products, together with standard deviations, are listed. The weighting spectrum is identical to the one used to weight the KFK-INR set |41|. Most of the standard deviations in table VII are rather small at intermediate neutron energies. The large number of isotopes causes a cancelling out of many of the errors. Large error

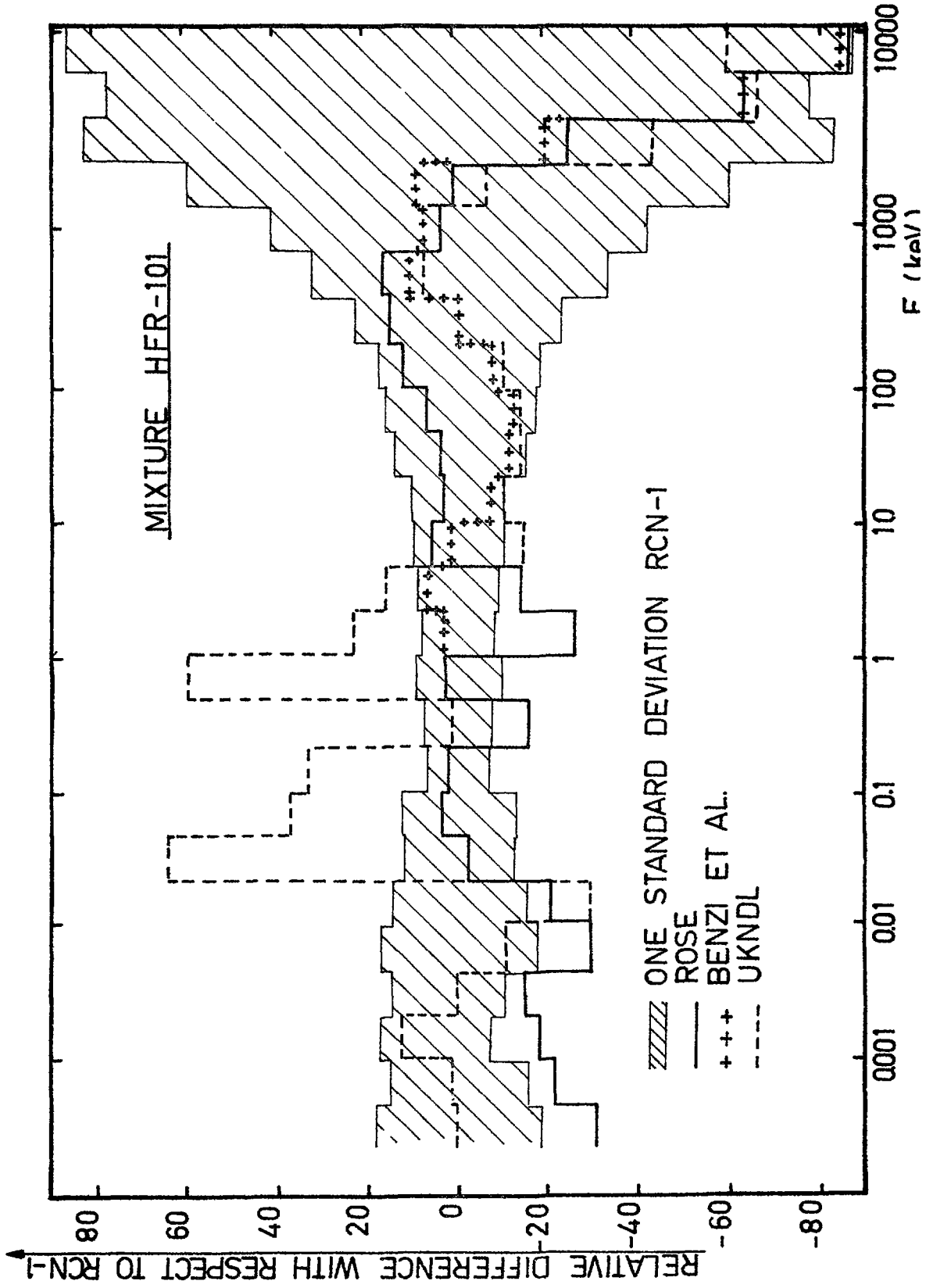


Fig. 5. Comparison of group cross sections for the HFR-101 sample for different sets.

Table VII Calculated capture cross sections for pseudo fission product mixtures from RCN-1 set in barns/fission (relative standard deviations (%) in parentheses).

energy group	^{235}U		^{238}U		^{239}Pu		^{241}Pu	
	calculated a priori	adjusted	calculated a priori	adjusted	calculated a priori	adjusted	calculated a priori	adjusted
1	0.011 (87)	0.009 (96)	0.014 (88)	0.011 (100)	0.016 (88)	0.012 (103)	0.017 (88)	0.013 (105)
2	0.016 (78)	0.013 (85)	0.021 (79)	0.017 (88)	0.023 (78)	0.019 (89)	0.025 (79)	0.020 (91)
3	0.024 (80)	0.020 (88)	0.031 (78)	0.026 (87)	0.035 (77)	0.028 (87)	0.038 (77)	0.030 (88)
4	0.037 (57)	0.033 (59)	0.047 (51)	0.042 (53)	0.053 (49)	0.046 (51)	0.057 (48)	0.049 (51)
5	0.056 (38)	0.053 (37)	0.072 (34)	0.066 (34)	0.080 (34)	0.072 (34)	0.086 (33)	0.077 (33)
6	0.086 (30)	0.081 (29)	0.107 (28)	0.100 (27)	0.119 (28)	0.109 (28)	0.128 (28)	0.116 (28)
7	0.13 (22)	0.13 (20)	0.16 (21)	0.15 (20)	0.18 (21)	0.17 (20)	0.20 (22)	0.18 (21)
8	0.19 (17)	0.19 (15)	0.25 (16)	0.23 (15)	0.28 (17)	0.26 (15)	0.30 (17)	0.27 (15)
9	0.30 (15)	0.30 (13)	0.38 (15)	0.37 (13)	0.43 (16)	0.40 (14)	0.47 (16)	0.43 (14)
10	0.47 (14)	0.46 (12)	0.59 (14)	0.57 (12)	0.67 (15)	0.62 (13)	0.72 (15)	0.65 (13)
11	0.70 (10)	0.70 (9)	0.87 (11)	0.85 (9)	0.97 (12)	0.91 (10)	1.04 (12)	0.96 (10)
12	1.03 (10)	1.03 (8)	1.27 (10)	1.24 (9)	1.37 (11)	1.31 (9)	1.46 (11)	1.37 (9)
13	1.51 (9)	1.53 (8)	1.85 (9)	1.84 (8)	1.98 (9)	1.92 (8)	2.10 (9)	2.01 (8)
14	2.50 (8)	2.55 (7)	3.04 (8)	3.04 (7)	3.18 (8)	3.13 (7)	3.37 (8)	3.27 (7)
15	3.9 (10)	4.0 (9)	4.8 (10)	4.8 (9)	5.0 (10)	4.9 (9)	5.5 (10)	5.4 (9)
16	8.6 (8)	8.7 (8)	10.0 (8)	10.0 (8)	10.1 (8)	10.0 (8)	10.4 (9)	10.3 (9)
17	10.7 (7)	10.6 (7)	12.7 (8)	12.6 (7)	12.8 (7)	12.7 (7)	13.7 (8)	13.5 (8)
18	11.0 (11)	10.8 (11)	15.3 (12)	14.9 (12)	17.0 (12)	16.7 (12)	19.4 (12)	19.1 (12)
19	28 (14)	27 (14)	33 (14)	33 (14)	34 (14)	34 (14)	38 (14)	37 (15)
20	40 (16)	40 (16)	50 (16)	50 (16)	52 (16)	52 (16)	45 (16)	45 (16)
21	77 (18)	77 (18)	107 (17)	106 (17)	110 (17)	110 (17)	132 (17)	131 (17)
22	24 (19)	23 (20)	31 (21)	29 (22)	29 (21)	28 (22)	33 (21)	32 (21)
23	34 (18)	33 (19)	61 (18)	59 (19)	59 (19)	58 (19)	57 (19)	56 (19)
24	78 (15)	75 (15)	131 (15)	125 (16)	106 (16)	103 (16)	113 (17)	109 (17)
25	43 (24)	41 (24)	68 (25)	64 (26)	66 (24)	63 (25)	73 (24)	70 (25)
26	699	692	1205	1193	1107	1098	1244	1234

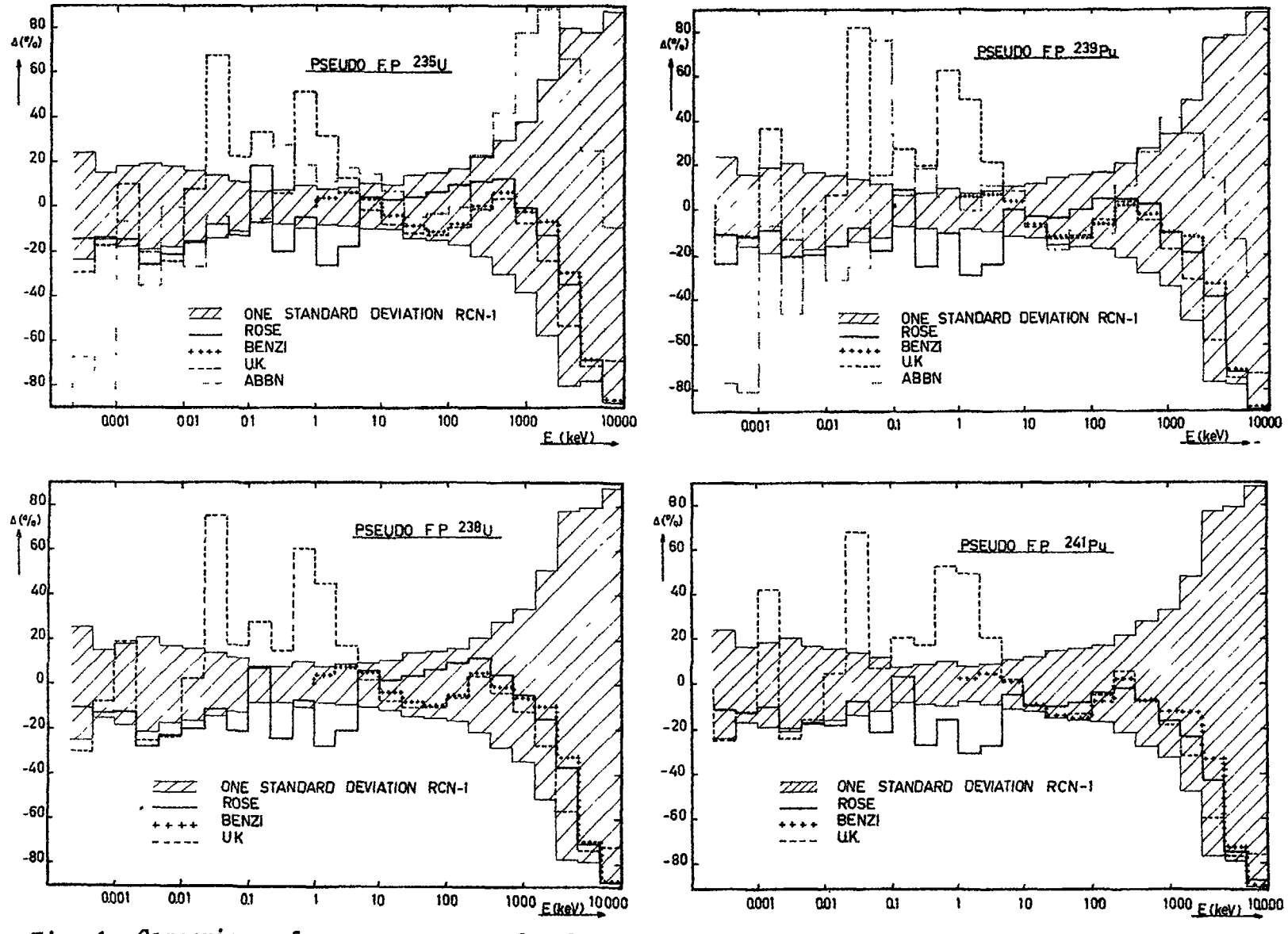


Fig. 6. Comparison of group constants for four pseudo fission product mixtures calculated with different cross section sets.

Plotted is the relative difference with respect to RCN-1 (%).

contributions are due to systematic model errors (at high energies) and (at low energies) the assumed weighting spectrum errors varying from 5% to 15% for the groups 15 to 26 |35,45|. The errors in group constants are strongly correlated.

In fig. 6 the RCN-1 set for the four pseudo fission products is compared with some other sets. It has to be noted that these sets are not always independent of each other. The sets ABBN and UKNDL show systematic deviations from RCN-1. In the latter case this is to a large extent due to low energy resonances of some isotopes in UKNDL which are not found in literature. The rather low values of ABBN cross sections at low energies are due to the circumstance that the contribution of some strongly absorbing isotopes are not included in this set for energies below 1 keV.

The group constants of the different mixtures are in general strongly correlated, since different mixtures have many isotopes in common. All correlation coefficients between cross sections for the 26 groups and the seven mixtures (four pseudo fission products and three integral samples) have been calculated. This is necessary for the application of the adjustment method (see section 3.2.1.4.).

Reaction rates and reactivity worths of pseudo fission product mixtures in the centre of the SNR-300, calculated with different cross section sets, are compared in table VIII. No errors have been assumed in the spectrum of SNR-300 or in the isotopic concentrations of the fission products in the mixtures. Standard deviations of at most 10% are obtained according to the RCN-1 error calculations. Deviations from results obtained with other sets are consistent with the calculated errors.

3.2.1.3. Analysis and conclusions of the FR 0 measurements on the KFK fission product mock-up sample. The results of the FR 0 measurements on the KFK fission product mock-up sample are summarized in table IX. The experimental data for the reactivity worths of samples of finite size have been transformed by us to data for infinitely thin samples by use of the published sample size correction factors |24|. In table IX, also results calculated by us with other cross section sets are given. In these calculations it is assumed that the composition of the KFK mock-up samples used in FR 0 is the same as that of the KFK mock-up samples used in STEK. The calculations were normalized to the values for the ABBN cross section set reported in |24|. From the table it is clear that, as in the case for the STEK measurements, the Australian set leads to the lowest data, while the UKNDL set produces the

Table VIII Calculated integral quantities of pseudo-fission product mixtures in SNR-300 for different cross section sets [a].

mixture	capture rate per fission per s ($\times 10^9$)				
	RCN-1 [33]	ABBN [34]	UKNDL [36]	Australian [31]	Benzi et al. [37]
^{235}U	1.96± 9%	2.19	2.08	1.95	1.94
^{238}U	2.43±10%	-	2.66	2.38	2.42
^{239}Pu	2.65±10%	2.64	2.89	2.47	2.57
^{241}Pu	2.84±10%	-	3.06	2.49	2.70

mixture	reactivity due to capture (arbitrary units)					
	RCN-1 [33]	ABBN [34]	UKNDL [36]	Australian [31]	Benzi et al. [37]	RCN-1 adj.
^{235}U	0.905± 9%	1.02	0.973	0.898	0.898	0.90±7%
^{238}U	1.12± 9%	-	1.24	1.09	1.12	1.09±8%
^{239}Pu	1.22±10%	1.23	1.35	1.13	1.19	1.16±8%
^{241}Pu	1.31±10%	-	1.43	1.14	1.25	1.22±8%

a) The sets RCN-1, UKNDL and Benzi have been supplemented, if necessary, with cross sections from the most complete set, which is the Australian set. At energies below 1 keV Benzi's set has been extended with the RCN-1 cross sections.

highest data, the RCN and Benzi set being in between.

Only the measurements in the soft spectrum core (core 5) are in some agreement with the data obtained from the RCN and Benzi sets. All other experimental values are much lower than the calculated ones. The disagreement increases with increasing hardness of the neutron spectrum. This tendency can also be seen in the STEK-1000 results.

No explanation for this discrepancy can be offered. A slight, undetected, amount of water absorbed in the sample would produce a trend like the observed one. In the STEK measurements such an disturbance has actually been observed [42]. To explain the deviation between the measured worths in core 8 and 3 and the worths calculated e.g. from the RCN-1 set, an amount of about 2.5 w/o water would have to be assumed, a rather high amount which is not likely to occur unobserved.

Table IX Comparison of experimental and calculated reactivity worths in FR 0 for the KFK mock-up sample.

The reactivity effect is expressed as the ratio of the effect of one pair of fission products to the effect of one atom of ^{235}U.

core	experiment	f a)	ABBN b)		RCN		Austral. c)	UKNDL c)	Benzi c)
			total	scatt.	total	scatt. c)			
3	-.043±.008	.998	-.156	-.025	-.118	-.015	-.116	-.114	-.115
8	-.103±.01	.986	-.232	-.020	-.172	-.013	-.162	-.180	-.170
8/Cu d)	-.122±.01	.986	-.229	-.020	-.171	-.008	-.158	-.180	-.169
5	-.401±.021 -.343±.007	.949 .941	-.501	-.011	-.388	-.008	-.323	-.412	-.388
5/Cu d)	-.298±.021 -.298±.007	.949 .941	-.459	-.011	-.358	-.005	-.303	-.382	-.358

a) Calculated finite sample size correction factor (see section 2.2.1.).

b) Cross section for a pair of fission products of ^{239}Pu were used; values given here are taken from [24].

c) Scattering contribution calculated from the Australian set.

d) Measurements with copper lining around the sample position (see section 2.2.1.).

The scattering contribution to the total reactivity effect is even in the case of core 3 so small that a serious error in the calculation of the scattering contribution is also not sufficient to explain the discrepancy.

3.2.1.4. Cross section adjustment.

The principle

The methods of cross section adjustment constitute a link between integral measurements and evaluated cross section data. The methods have mostly been applied until now for fissile and construction materials of fast reactors. However, the application to fission product data is promising. The adjustment techniques may be considered from two different points of view.

a. Certain margins are set for the cross sections (usually in fairly coarse energy groups). It is tried to change the cross sections within these margins in such a way that an optimal agreement is reached between the calculation and the measurement of integral quantities. The optimalization may be performed, e.g. by a least squares technique. This may often lead to improvements of the group cross sections, and, in any event to improvement in the prediction of integral parameters of a power reactor.

b. In order to predict some integral effect in the best possible way an optimal combination of integral measurements and evaluated different data can be sought. A minimum variance criterion may be shown to lead to a method which is mathematically equivalent to the least squares adjustment |43|. This way of interpreting the adjustment may be favoured in cases where the adjustment bearly means an improvement of the cross sections, like the adjustment of pseudo fission product cross sections based on measurements with fission product mixtures (see below).

Cross section margins

The above mentioned cross section margins correspond with the uncertainties, say the standard deviations. It is important - especially if the number of data is large - to take into account the correlations that exist between the cross sections at different energies and between cross sections of different isotopes. At RCN an attempt has been made to calculate the uncertainties for a number of fission product isotopes and mixtures, starting from first principles |35|, for the RCN-1 cross section set |33|. The following error sources have been taken into account to calculate the (co)variances: errors in all (resonance) parameters used in the cross section calculations; uncertainties in nuclear level statistics and Porter-Thomas width fluctuations; model errors and uncertainties in the group weighting spectra.

Systematic errors

A hazard in the application of adjustment techniques is the presence of systematic errors in the integral data. The following example illustrates what may happen. Suppose that the reactivity effect is measured in a rather soft spectrum of an isotope with fairly accurately known low energy cross sections (well known resolved resonance data) and larger errors above 1 keV. Suppose that selfshielding occurs at low energy, which remains unnoticed and is not taken into account. The reactivity worth is then overpredicted. If now an adjustment technique is applied, the cross section will be lowered, especially so above 1 keV,

where the largest margins occur. If the adjusted data are used subsequently to calculate some effect in a fast breeder, where no neutrons below 1 keV occur, the result will be too low. It may happen that a small error in the experiment may be amplified to a large error in the fast breeder effect. Such an effect has been found in practice at RCN |44| for measurements on neutron capture in ^{103}Rh in some STEK cores. It was observed that a systematic error of 20% in the selfshielding. could be amplified to an error of a factor of 2 in the capture rate in SNR-300. The application of statistical tests is essential to recognize possible systematic errors.

Adjustment of pseudo fission product capture cross sections

In ref. |45| the adjustment technique has been applied to improve group cross sections of four pseudo fission products using reactivity worths of three different integral fission product mixtures, measured in four different spectra in STEK. From the error analysis, mentioned before, the complete covariance matrix was obtained of the cross section vector, consisting of the 26 group constants of the 7 mixtures (3 measured integral samples and 4 pseudo fission products). It may be noted that the group constants of the different mixtures are in general strongly correlated, since different mixtures have many isotopes in common. If the integral sample part of the cross section vector is adjusted to fit the experimental reactivity worths mentioned in table V the correlations will cause the cross sections of the pseudo fission products to follow this adjustment. In this way the differences in composition between integral samples and pseudo fission products are taken into account. The experimental information is then used to an optimum in improving the pseudo fission product cross sections.

The adjusted data obtained in this way are used to derive the data for the three integral samples for four STEK cores given in table VI. They are also given for the pseudo fission products in table VII.

The accuracy of the group cross sections is not improved noticeably. A somewhat larger improvement is obtained when some integral quantities of a fast breeder are calculated with the adjusted data, as given in table VIII. Nevertheless, the improvement is still small. This is due to the fact that remarkably low estimates have been obtained for the standard deviations of the calculated group cross sections, due to cancellation of many errors in the mixtures of many isotopes.

It may finally be mentioned that in this situation the χ^2-test, including all correlations in experimental errors, gave no indication

of systematic errors.

3.3.2. Results of measurements on isotopic samples

With regard to the isotopic sample measurements a complete analysis has been made and published only for the FR 0 measurements.
Except for a few isotopes the STEK measurements have not yet been analyzed in detail. The main problem is still the correction for selfshielding and to a lesser extent flux depression. Up to now only a simple extrapolation to zero sample mass has been made for most of the samples. Calculations with more sophisticated methods are underway. On the activation and reactivity worth measurements in the CFRMF only some, partly preliminary, results are available.
The results of the measurements in three FR 0 cores have been compared by us with calculated values obtained from several cross section sets. In this comparison use was made of 26 group cross sections derived from the different data files using a STEK standard weighting spectrum, i.e. a smoothed STEK-2000 spectrum. This weighting will not always be appropriate for all cores but the effect, except for very special cases, is not of great importance for the comparison.
In all cases the experimentally obtained values are transformed into values for infinite thin samples by use of the published sample size correction factors. In the comparison of other sets with the FR 0 data the calculations are normalized to the published data obtained from the AE SPENG set |24|. Also preliminary data from CFRMF and some data from STEK are taken into account in this comparison. The result of the comparison is given in table X. In the table is given the ratio of the calculated effect (C) to the measured effect (E). These effects include the scattering contribution. For the calculations with the RCN-1 |33|, Australian |31|, UKNDL |36| and Benzi |37| sets we have always used the scattering matrices from the Australian set. For energies below 1 keV the Benzi set is supplemented with RCN-1 group constants.
The scattering contributions calculated with the Australian data are always larger than those calculated from the scattering matrices included in the SPENG set, sometimes up to a factor of 2.
Since the scattering contribution for the isotopes considered is only occasionally larger than 10%, it usually being much smaller, the difference in scattering contributions as calculated from the different sets, will not be of great importance for the comparison.

Table X

Comparison of results of experiments (E) and calculations (C) for different group sets for a number of infinitively small isotopic samples, measured in FR 0, STEK and CFRMF.

^{95}Mo

measurement	exp. [a]	f	C/E				
			SPENG	RCN-1	Austral.	UKNDL	Benzi
FR 0							
3	-0.075±7%	0.991	1.07	1.11	1.15	1.07	1.04
8	-0.133±6%	0.860	1.0	0.97	0.93	0.97	1.10
5	-0.293±3%	0.590	1.0	1.0	0.92	1.0	0.97
CFRMF react. worth	-0.22 [b]	-	-	0.84	0.87	0.91	0.76

^{97}Mo

measurement	exp. [a]	f	C/E				
			SPENG	RCN-1	Austral.	UKNDL	Benzi
FR 0							
3	-0.077±7%	0.992	1.15	1.28	1.0	1.21	1.21
8	-0.111±6%	0.934	1.28	1.18	0.94	1.21	1.12
5	-0.150±4%	0.863	1.50	1.17	1.01	1.32	1.16
CFRMF react. worth	-0.25 [b]	-	-	0.60	0.52	0.64	0.64

^{99}Tc

measurement	exp. [a]	f	C/E				
			SPENG	RCN-1	Austral.	UKNDL	Benzi
FR 0							
3	-0.118±17%	0.996	2.0	1.66	1.09	1.55	1.62
8	-0.188±13%	0.960	1.56	1.44	0.97	1.36	1.57
5	-0.418±5%	0.774	1.23	1.29	0.81	1.06	1.45
CFRMF $\bar{\sigma}_{act}$ [b]	0.294±13%	-	-	1.42	0.88	1.36	1.53
ERMINE	-	-	-	-	0.9±.2	1.2±.2	

Discussions of the comparison and some conclusions

^{95}Mo: It is seen that the results of all sets are in rather good agreement with each other and with the experiments. Apparently the RCN-1 and Australian set overpredict the absorption at higher energies, while the Australian set underpredicts the absorption at low energies. Preliminary results of the STEK-cores 1000 and 2000 with relative hard neutron spectra (see fig. 7) seem to support the good prediction ability of all sets. Preliminary results of STEK-cores 3000 and 4000 with soft spectra, however, fail to agree which may be caused by a gross underestimation of the selfshielding correction, still to be calculated in more detail.

The reactivity worth measurements in the CFRMF seem to be underpredicted by all sets. However, in this case the scattering contribution is very large (about half of the total effect) and no definite conclusion can thus be drawn yet.

^{97}Mo: Here in general an overprediction is observed. The Australian set seems to give a correct prediction. All other sets lead to an overprediction. The same impression is obtained from some STEK measurements for which also the SPENG set leads to an overprediction and the Australian set to an underprediction. The behaviour of the other sets for STEK resembles that for FR 0.

The CFRMF reactivity worth measurement is about a factor two lower predicted by all sets which seem in contradiction to the FR 0 core 3 results.

^{99}Tc: The SPENG set apparently leads to a large overprediction as do also the RCN-1, UKNDL and Benzi sets. Again only the Australian set gives a lower prediction. In particular should be noted here that the overprediction increases with increasing hardness of the spectrum indicating too large a high energy capture. This is, however, not supported by preliminary STEK results, which, even allowing for gross errors in the estimated effect of selfshielding, show an opposite trend.

From the CFRMF activation results might also be concluded to an overprediction for higher energies for most sets.
The preliminary ERMINE results support the above general conclusions.

^{101}Ru: The SPENG set leads to a moderate overprediction for all cores, while the RCN-1 set for cores 8 and 5 produce better values. Striking is the magnitude of the overprediction of the Australian set, which is caused by the high $\sigma_{n\gamma}$ above 3 keV, in this set. In general, the Benzi set (supplemented with RCN-1 group constants for energies

Table X (continued)

^{101}Ru

measurement	exp. [a]	f	C/E				
			SPENG	RCN-1	Austral.	UKNDL	Benzi
FR 0							
3	-0.17 ±10%	0.989	1.15	1.31	3.4	0.97	1.12
8	-0.276±7%	0.905	1.19	1.02	2.2	1.04	1.03
5	-0.464±4%	0.797	1.26	0.90	1.46	1.38	1.00

^{102}Ru

measurement	exp. [a]	f	C/E				
			SPENG	RCN-1	Austral.	UKNDL	Benzi
FR 0							
3	-0.049±23%	0.996	2	2.70	2.70	2.01	2.07
8	-0.061±5%	0.880	2.46	1.82	1.82	2.39	2.10
5	-0.105±15%	0.704	3.33	1.63	1.45	3.12	1.77
CFRMF			HEDL [28]				
$\bar{\sigma}_{act}$ [b]	0.097±11%	-	1.3	1.65	1.68	2.4	2.1
ERMINE	-	-	-	-	> 1	> 1	-

^{104}Ru

measurement	exp. [a]	f	C/E				
			SPENG	RCN-1	Austral.	UKNDL	Benzi
FR 0							
3	-0.059±30%	0.997	0.71	1.4	0.65	0.84	0.79
8	-0.047±40%	0.993	1.21	2.3	1.19	2.19	1.30
5	-0.095±17%	0.982	1.01	1.51	0.86	3.94	1.23
CFRMF			HEDL [28]				
$\bar{\sigma}_{act}$ [b]	0.932±10%		0.90				

^{103}Rh

measurement	exp. [a]	f	C/E				
			SPENG	RCN-1	Austral.	UKNDL	Benzi
FR 0							
3	-0.177±5%	0.992	1	0.93	0.98	1.04	1.03
8	-0.279±8%	0.906	0.93	0.81	0.81	1.13	0.91
5	-0.95 ±3%	0.552	0.69	0.62	0.59	0.89	0.65

below 1 keV) seems in agreement with the experiments. The STEK measurements tend to favour the RCN-1 set; the other sets, except the Benzi set, tends to an overprediction.

^{102}Ru: All sets lead to gross overpredictions of a factor of about 2. The same may be concluded from some STEK measurements where the results for different sets differ still more, especially in the soft spectra.

The CFRMF activation measurement is also overpredicted. The best value here is that of ref. |28|.
Apparently the cross sections of ^{102}Ru are seriously in error.

^{104}Ru: Taking into account the large error margins in the experimental data, especially in cores 3 and 8, the SPENG, Australian and Benzi sets lead to acceptable predictions. The RCN-1 set is generally too high, which is particularly the case for the UKNDL set for softer spectra. The same trends can be deduced from STEK measurements where especially the UKNDL set leads to very large overpredictions.

The CFRMF activation measurements show a good agreement with predictions based on the Benzi set and the HEDL data. The RCN-1 set is too high as is the UKNDL set, which seems in contradiction to the FR 0 core-3 data for the UKNDL set.

^{103}Rh: Due to important resonances at low energies the selfshielding of a Rh sample in a soft neutron spectrum may be quite large and thus the correction may be somewhat uncertain. This may be the reason for the systematic underprediction for the FR 0 core-8 data by all sets.
Otherwise the predictions agree rather well with the measurements.
For the STEK measurements of which some results are given in the table, the selfshielding has been calculated by the TRIX-code. Here the SPENG set seems to give predictions closest to the experimental data.
The RCN set, the Australian set and the Benzi set are consistently underpredicting, only the UKNDL set leads to an overprediction.
When an earlier value of 0.92 for the activation ratio of ^{103}Rh to ^{197}Au in CFRMF is combined with a value of 478 mb for the averaged activation cross section of ^{197}Au in CFRMF, calculated in |36|, a value of 450 mb is found for the $\bar{\sigma}_{act}$ of ^{103}Rh in CFRMF. This value is underpredicted by all sets, which is in agreement with the underprediction found for STEK. The preliminary results of ERMINE support the general trend.

^{133}Cs: In the FR 0 measurements two compounds were used, CsF and Cs_2CO_3. Only the data for the CsF are used here because the Cs_2CO_3 data are probably less reliable. For the hard spectrum in core 3 an overprediction is observed for all sets. This overprediction decreases or becomes an

Table X (continued)

^{103}Rh (continued)

measurement	exp. a)	f	C/E				
			SPENG	RCN-1	Austral.	UKNDL	Benzi
STEK-1000	-.46 ±3%	0.98	0.94	0.76	0.70	1.27	0.87
STEK-2000	-.52 ±2.5%	0.96	0.95	0.76	0.68	1.37	0.87
STEK-3000	-.60 ±4%	0.77	0.97	0.79	0.72	1.39	0.87
STEK-4000	-1.16±4%	0.88	0.90	0.80	0.77	1.17	0.83
CFRMF $\sigma_{act.b}$	0.450±?			0.74	0.73	0.79	0.83
ERMINE	-	-	-	-	0.9±.1	1.3 ±.1	-

^{133}Cs

measurement	exp. a)	f	C/E				
			SPENG	RCN-1	Austral.	UKNDL	Benzi
FR 0							
3	-0.087±8%	0.991	1.6	1.60	1.17	1.24	1.62
8	-0.162±6%	0.841	1.31	1.33	1.01	1.30	1.43
5	-0.472±2.4%	0.534	1.06	1.02	0.89	1.09	1.08
STEK-1000	-.42 ±3%	0.91	0.80	0.83	0.60	0.87	0.94
STEK-2000	-.59 ±6%	0.90	0.80	0.80	0.60	0.89	0.90
STEK-3000	-.62 ±6%	0.85	0.84	0.83	0.70	0.95	0.93
STEK-4000	-.94 ±6%	0.71	0.83	0.78	0.70	0.86	0.82
CFRMF $\bar{\sigma}_{act.b}$	0.336±8%	-	-	0.96	0.68	0.86	1.01

^{147}Pm

measurement	exp. a)	f	C/E				
			SPENG	RCN-1	Austral.	UKNDL	Benzi
FR 0							
3	-0.142±30%	0.995	1.75	1.54	1.65	1.80	1.44
8	-0.407±14%	0.900	1.60	1.19	1.19	1.13	1.12
5	-1.14 ±5%	0.778	1.75	1.50	1.48	1.05	1.45
CFRMF $\bar{\sigma}_{act.b}$	0.841±9.5%	-	-	0.84	0.84	0.84	-

underprediction going to the softer spectra. The Australian data seem here to lead to a prediction closest to the experiment. In the case of the STEK data, analyzed by calculating the selfshielding by the TRIX code, the prediction fluctuates much less going from hard to soft spectra. Remarkable is here the consistent underprediction produced by all sets, the Australian set being lowest and far outside the experimental error limits. The best set here seems the Benzi set.

The CFRMF activation measurement is also supported by the Benzi set and also by the RCN-1 set. This seems in contradiction to the FR 0 core 3 results.
It might be that the CsF sample contained some undetected moisture. An amount of about 2.5 w/o would be needed to cancel the overpredictions in cores 3, 5 and 8 for the SPENG set.

^{147}Pm: It is not clear why the prediction for different cores and different sets scatter so much. The overprediction in core 3 is contradicted by the underprediction of the activation measurement in CFRMF. The change in the prediction trend in core 8 for the RCN-1, Australian and Benzi sets needs a further careful study.
Some preliminary STEK data seem as far as they have been analyzed, more to point to underpredictions for all STEK cores by all sets, except the SPENG set. But due to the great uncertainty still existing for the selfshielding corrections no definite conclusions can be drawn, as yet.

^{149}Sm: In all cores the effect is underpredicted. The SPENG set produces the best data, followed by the Benzi set, UKNDL set, Australian set and the RCN-1 set. The change in trend going from hard spectrum to soft spectrum might be caused by an error in the selfshielding correction, in the soft spectrum case this correction being quite large.
The underprediction is also observed for the CFRMF reactivity worth measurement, however, the rate of underprediction is larger.

^{127}I: This isotope has not been measured in FR 0. For the STEK measurements an analysis has been made using TRIX calculations. From this it is seen that in all sets, except UKNDL, the high energy cross sections seem too small, the Benzi set being the best one.
The UKNDL set overpredicts rather much.

There are two CFRMF measurements. The result of the reactivity worth measurement is rather well predicted by all sets except by the UKNDL. The result of the activation measurement is slightly more overpredicted. The error quoted in this measurement does not include the uncertainty resulting from the problems associated with the decay scheme of ^{128}I.

Table X (continued)

^{149}Sm

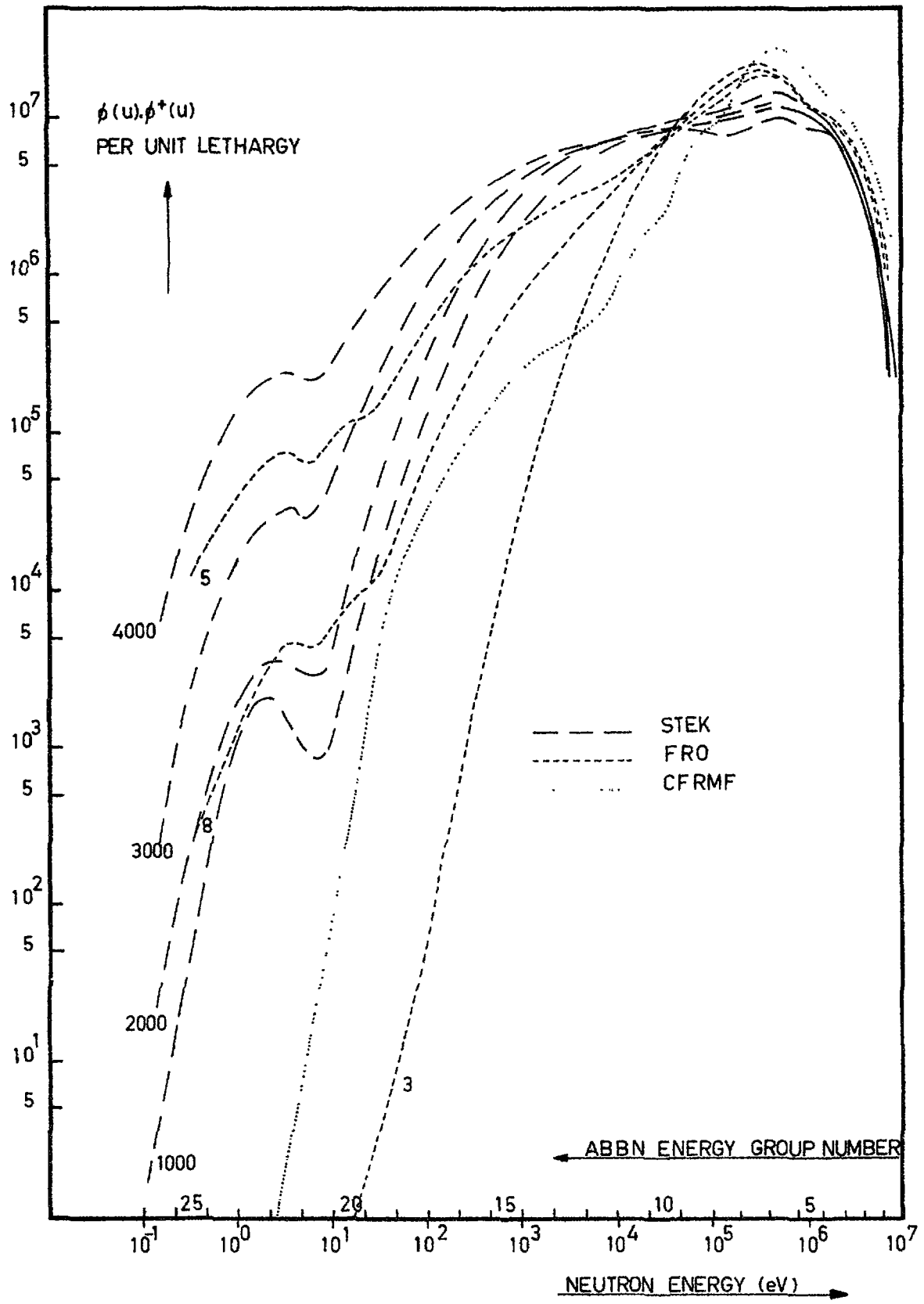
measurement	exp. [a]	f	C/E				
			SPENG	RCN-1	Austral.	UKNDL	Benzi
FR 0 3	-0.436±26%	0.99	0.83	0.66	0.73	0.80	0.87
8	-0.919±11%	0.799	0.98	0.78	0.63	0.84	0.85
5	-4.07 ±3%	0.504	0.86	0.60	0.60	0.68	0.82
CFRMF react. worth	-0.58 [b]	-	-	0.65	0.56	0.74	0.75

^{127}I

measurement	exp. [a]	f	C/E				
			ENDF/B IV [49]	RCN-1	Austral.	UKNDL	Benzi
STEK-1000	-.50 ±12%	0.86	0.71	0.73	0.62	1.45	0.80
STEK-2000	-.65 ±10%	0.78	0.71	0.73	0.61	1.50	0.77
STEK-3000	-.75 ±10%	0.77	0.83	0.84	0.75	1.	0.88
STEK-4000	-.89 ±7%	0.68	0.93	0.94	0.88	1.53	0.96
CFRMF react. worth	-0.20 [b]	-	1.00	1.08	1.02	1.27	1.11
$\sigma_{act.b}$	0.304±9%	-	HEDL [28] 1.07	1.17	1.05	1.78	1.23

a) The experimental reactivity worth quantities are:
- for FR 0 the effect of one atom of the isotope to the effect of one atom of ^{235}U;
- for STEK the quantity ρ_{sample}/ρ_0 as defined in section 2.3;
- for CFRMF the ratio of the reactivity effect of a gram of the isotope considered to the reactivity effect of a gram ^{235}U.

b) Estimated values, including tentative corrections calculated by us.



VALUES OF $\phi(u).\phi^+(u)$ FOR DIFFERENT CORES REVIEWED

FIG.7

4. CONCLUSIONS AND RECOMMENDATIONS

There is only one measurement of thermal capture of gross fission products, i.e. the combined Canadian-Swedish measurement in R0. No data for resonance capture is given.

The accuracy of the thermal capture measurement in 5%, the over-prediction by FISSPROD calculations is 7%. Thus the situation is not really satisfying. The forthcoming analysis of the CEA experiments with irradiated SENA fuel pins measured in Minerve-Melodie (see appendix) might produce slightly more accurate data. The main error sources are the uncertainties in the amounts of residual fuel in the gross fission product samples. Unless one succeeds in reducing these uncertainties to less than a few tenths of a percent or is able to design a system or a measuring technique in which the residual fuel has much less influence, the accuracy of future integral measurements of thermal absorption gross fission products by the reactivity worth measurement of irradiated fuel samples, will not be much better than the 5% presently reached.

When the fast gross fission product absorption as measured in STEK is checked against calculations with the RCN-1 set deviations ranging from +14% to -24% are observed. Here also the uncertainty in the amount of residual fuel is one of the sources for the limited accuracy, however not as overriding as in the thermal case. Other sources for the deviations are to be found in uncertainties in the neutron spectrum, in the scattering and in the selfshielding in the samples.

Reactivity effects of pseudo fission products in SNR-300 when calculated with the RCN-1 adjusted set, have standard deviations of the order of 10% as far as cross section uncertainties are concerned. Uncertainties in the yields and spectrum have to be added, however. The STEK measurements cover a lower energy range than the ERMINE measurements (see appendix). Moreover most fission products used in the ERMINE measurements are generated by fast fission in ^{239}Pu, thus yield uncertainties will have a smaller influence in an analysis directed to the generation or checking of pseudo-fission products for fast reactors. It will be most interesting to combine the forthcoming ERMINE results with the STEK results in one adjustment procedure. Conclusions on further measurements on gross fission product samples in fast spectra have to wait until this combined analysis has been made.

From the isotopic sample measurements only some preliminary conclusions will be drawn here pending the further analysis of the STEK and CFRMF measurements and of the completion and analysis of the measurements in progress in ERMINE.
When all these data are available and compared (e.g. in a year's time) a much better picture of the situation will be obtained. However, already now it can be concluded that apparently ^{99}Tc, ^{102}Ru and ^{147}Pm are overpredicted by all or nearly all sets, while ^{149}Sm is always underpredicted. A new evaluation of these isotopes seems necessary. In comparing the different sets it is found that the UKNDL set and the Australian set deviate in general more from the experimental values than the SPENG, RCN-I and Benzi sets do. The Australian set has a tendency to be consistently low.

The error in the measurements including errors in the applied corrections appear to be of the order of 5% or more. However, in many instances the differences between data calculated from different sets are much larger than the experimental errors. Thus, integral data, even with a limited accuracy, will be of advantage in the evaluation of cross sections.

Further integral measurements on isotopic samples may be needed in future to clear up discrepancies in measurements. It may also prove necessary to extend the energy range to regions below that presently used in order to collect data applicable for very large LMFBR's with soft spectra.

The scattering which gives an important contribution to the reactivity worth in hard spectra for not very absorbing isotopes, has to be determined with greater precision. For this purpose special measurements with isotopes which mainly exhibit scattering can be made to check and improve the nuclear models used for the calculation of the scattering. Since scattering does not play any role in the activation and transmutation, this type of measurement should also be used at its greatest extent. In order to provide several different spectra for the irradiations, the use of spectrum shaping filters might be considered.

Finally, for the reactivity measurements, there is still the problem of lacking of good and reliable standard materials. The same holds for the activation and the transmutation measurements.

The extent to which the needs for data for individual fission products, as stated in table VI of review paper 3 are covered by the present measurements is given in table XI.

Table XI

Isotopes measured (or to be measured) in present integral programs arranged according to the list of needs as given in table VI of Review Paper 3.

Fission product group I

	FR 0 3 cores	CFRMF activ.	ERMINE react.	STEK 5 cores	ERMINE
Ru-101	X			X	X
Rh-103	X			X	X
Tc-99	X	X		X	X
Cs-133	X	X		X	X
Pd-105				X	X
Ru-102	X	X		X	X
Pd-107				X	X
Xe-131				X	
Nd-143				X	X
Pm-147	X	X		X	X
Sm-149	X		X	X	X
Sm-151				X	X
Mo-97	X		X	X	X

Fission product group II

	FR 0 3 cores	CFRMF activ.	ERMINE react.	STEK 5 cores	ERMINE
Mo-95	X			X	
Cs-135				X	
Nd-145				X	X
Mo-98			X	X	
Ag-109				X	X
Ru-104	X	X		X	
Eu-153		X	X	X	X
Pr-141		X		X	
Xe-132		X			
Mo-100			X	X	
Sm-152				X	
Eu-154					
Zr-93				X	

Table XI (continued)

Fission product group III

	FR 0 3 cores	CFRMF activ.	ERMINE react.	STEK 5 cores	ERMINE
Sm-147			X	X	
Nd-148		X		X	
Ru-103					
Pd-106				X	
La-139				X	
Nd-144				X	
Eu-155					
Nd-146				X	
Xe-134		X			
I-127		X	X	X	
Pd-104				X	
I-129				X	
Pd-108				X	

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Appendix 1

This review paper is confined primarily to those experiments on which results are now available. Besides those experiments mentioned there are some other activities which may, at some time, lead to additional integral data on the neutron capture in fission products.
Two of these activities will be summarized here.

The first concerns measurements of integral cross sections for heavy metals (mainly ^{240}Pu) and for fission products in HTR's [6]. These experiments are being undertaken jointly by the DRAGON project, CEA of France and the UKAEA. Fuel compacts of the "Teledial" type (12 mm diameter by 50 mm long) are being irradiated in DRAGON. After irradiation the reactivity controlled by these compacts will be measured in the reactor HECTOR at Winfrith and will be compared with that controlled by similar unirradiated compacts to give a direct measure of the effects of irradiation. It is planned to use these data to obtain a measure of the absorption of ^{240}Pu and fission products in the sample in a typical HTR spectrum. As an aid in correcting for the effects of other isotopes in the sample, measurements will also be made in a thermal spectrum.

In order to determine the composition of the irradiated fuel compacts extensive chemical and mass spectrometer measurements on them will be carried out in France.

It is planned to carry out the first set of measurements in HECTOR early in 1974 when samples irradiated to about 40.000 MWD/Te will be available. Later measurements in 1975 could extend this work to 80.000 MWD/Te samples, and it is hoped to determine absorptions in ^{240}Pu plus fission products to an accuracy of about ± 3.5% in the most highly irradiated samples.

As second activity can be mentioned the experiments of the CEA, France for light water moderated reactors as well as for fast reactors [47]. These experiments are primarily aimed at a global checking of the evolution of reactivity due to burnup and conversion, and the fission product formation and capture. In these experiments the amounts of the heavy metals in the irradiated fuel samples and their reactivity effects will be determined sufficiently accurately to enable also the determination of integral capture data for fission products. Furthermore some measurements in fast spectra on separated fission product isotopes will take place.

The reactivity worth measurements are performed on irradiated fuel pins in a critical reactor in a lattice position. This lattice is very much like the lattice of the reactor under study and in which the sample pins have been irradiated. Since the irradiated fuel pin occupies a normal fuel pin position in an otherwise unperturbed lattice, normal cell calculation codes can be used in the analysis and the results are also directly applicable to actual reactor systems.

The following experiments can be mentioned here:

a) For graphite natural uranium reactors. In the zero power reactors Marius (low temperature) and Cesar (high temperature) the reactivity changes in irradiated fuel rods have been determined. From these measurements it could be concluded that the capture of fission products is overestimated by about 5% |48|.

b) For the water reactor measurements a number of fuel pins have been (and are) irradiated in the SENA reactor at Chooz (PWR, 300 MWe). In the reactor Minerve at Fontenay-aux-Roses, three different test configurations have been foreseen consisting of 800 cells of the SENA type surrounded by a thermal driver consisting of the normal highly enriched plate type fuel elements of Minerve. The fuel of the SENA cells consists of pins of uranium oxyde and mixed uranium plutonium oxydes. Under an agreement of collaboration this fuel was obtained from the CEN at Mol, where it has been used in the zero power reactor Venus.

   The three configurations simulate three different burnup stages of the SENA reactor (15, 22 and 30 MWd/kg respectively). A number of irradiated fuel pins (10 cm long) with different burnup are available. A great number of pin samples is available with different heavy metal compositions for calibrations of the heavy metal effects. The effect of fission products is normalized on ^{235}U and boron by use of special UO_2 + B samples.

   The first series of measurements was concluded in 1972. A preliminary analysis shows that the effect of the gross fission products can be determined with an accuracy of 5% or even better. A comparison of the experimentally determined capture data with data derived from nuclear data files is now underway. Further measurements are foreseen for 1973 and 1974.

For fast reactor measurements the same principle is used as for the water reactors. Sample fuel pins are irradiated in Rapsodie and/or Phenix and are measured in ERMINE, the coupled thermal-fast system in Minerve. The program is aiming at an accuracy close to 5% in the experimentally determined capture of fission products. The program is composed of two phases. In phase I, realized in 1973, fuel sample pins of uranium-plutonium oxyde irradiated in Rapsodie to about 6 to 8% burnup (about 2/3 of the number of fissions from ^{235}U and 1/3 from Pu) are oscillated in ERMINE (V.R3) which has a central zone with 15% enriched ^{235}U as fuel and stainless steel sodium and oxygen as diluent. This core simulates a fast power reactor core. The samples contain 100-150 g fissile materials and 10 g of fission products. The reactivity worths of these samples are compared to the worth of unirradiated pins and of pins with different heavy metal compositions. Afterwards a destructive chemical analysis will provide the actual fissile isotope composition (accuracy around 0.1%) and fission product content (by measuring the Nd content up to a 2% accuracy). The accuracy of the measurement of the fission product effect is estimated to be better than 5%, the inaccuracy due to the chemical analysis may contribute to another 5% uncertainty in the fission product capture.
In phase II, to be started at the end of 1974 measurements in three cores in ERMINE are foreseen, simulating, also by use of depleted uranium and plutonium, cores of large power breeders.
As samples will be used
- pins of highly enriched uranium oxyde irradiated in Rapsodie;
- pins of depleted uranium-plutonium oxyde with a high plutonium content irradiated in Rapsodie;
- pins of depleted uranium-plutonium oxyde, normal Phenix fuel.

The burnup will be about 10%. The same type of reference samples as in phase I will be used. The measurements of phase I are finished and results will be available soon.

Finally, measurements on separated isotopes are in the process of being performed. These concern the isotopes, $^{95,97}Mo$, ^{99}Tc, $^{101,102}Ru$, ^{103}Rh, $^{105,107}Pd$, ^{109}Ag, ^{133}Cs, $^{143,145}Nd$, ^{147}Pm, $^{149,151}Sm$ and ^{153}Eu. Oscillation measurements on these isotopes have been performed in ERMINE (V.R3). In the framework of a collaboration CEA-RCN six isotopic samples measured in STEK have also been measured in ERMINE (V.R3).
In addition irradiation experiments are being performed on isotopic samples in Rapsodie-Fortissimo (experiment TACO) and in Phenix (irradiation PROFIL). The transmutation will be measured by mass spectrometry.

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Review Paper no. 15

RESIDUAL POWER DUE TO FISSION PRODUCTS

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Abstract:

Various evaluations of the residual power emitted by the fission products of the main fissionable nuclides are compared among themselves and with experimental results. The consistency of the evaluations, which is of the order of 10%, is generally better than that of the corresponding experiments. Efforts should be made to:

- improve the basic data, especially those on short-lived nuclides;
- estimate the errors in evaluation arising from the errors in the basic data;
- improve the integral experiments qualitatively and quantitatively.

1. INTRODUCTION

There have been surprisingly few integral experiments relating to the energy released by the fission products of irradiated fuel, despite the fact that this energy represents 5-6% of the power produced by fission and has a considerable influence on the emergency cooling, handling, transport and reprocessing of fuel. The importance of this subject has been confirmed by D.J. Horen and A.M. Weinberg [1], who noted in their introductory paper presented at the IAEA Symposium on Applications of Nuclear Data in Science and Technology (Paris, March 1973) that some 22 000 pages have been written just on the problem of core cooling after shut-down (emergency core-cooling systems)

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without exhausting it, for the engineer who reverts to this problem finds himself confronted with inconsistent experimental values, while calculations based on fission product data files are also inconsistent, the data for short cooling times being incomplete.

In the first part of this paper we present a comparison of residual power evaluations which highlights the ranges of cooling times where there is agreement and disagreement. In the second part we compare theoretical evaluations with experimental results.

2. COMPARISON OF EVALUATIONS

2.1. Inquiry carried out in connection with this Panel

In order to obtain an objective picture of the consistency of the residual power values used in different reactor design projects, an inquiry was carried out in connection with this Panel among a fairly large number of laboratories and among the contributors mentioned in Paper 15. The following questions were asked:

(a) Residual power after one fission for ^{235}U th, ^{239}Pu th, ^{233}U th, ^{235}U fast, ^{239}Pu fast;

(b) Residual power after irradiation for one year at one fission event per second and without capture for the same fissionable nuclides.

In both cases the cooling times considered lay between 10 sec and 5×10^8 sec.

Unfortunately, only three authors gave precise answers, which limits the value of the inquiry and has led us to fill out the comparison with published values of which some are old and relate to only a limited number of cases about which the information reported here is incomplete and perhaps erroneous.

The following evaluations are compared among themselves:

Tasaka and Sasamoto, contribution to Paper 15 [2]

Vossebrecker, contribution to Paper 15 [3]

Devillers et al., contribution to Paper 15 [4]

Battat, Dudziak and Hicks [5]

Perkins [6]

Tobias [7]

England [8]

Sporrer [9]

Shure [10] (this evaluation is particularly interesting as the American Nuclear Society proposed it as a standard evaluation in 1971 [11]).

2.2. General description of the evaluations

The first theoretical evaluation of residual power was made by Way and Wigner [12], who proposed the formula $P = At^{-0.2}$ for the power released as a function of the time t after the end of infinite irradiation.

Since then, two main approaches have been adopted:

(a) The first, based mainly on a set of experimental results, led Shure to propose an analytical representation for the power P following infinite irradiation at power P_o. Shure's formula was taken by the American Nuclear Society as a standard formula for the energy released in water-cooled and -moderated reactors:

$$\frac{P}{P_o}(\infty, t_s) = At_s^{-a}$$

where $\frac{P}{P_o}(\infty, t_s)$ is the fraction of the power during operation, t_s is the cooling time, and A,a are given by the following table:

	A	a
$10^{-1} < t_s < 10^{1}$	12.06	0.0639
$10^{1} < t_s < 1.5 \times 10^{2}$	15.32	0.181
$1.5 \times 10^{2} < t_s < 4 \times 10^{6}$	26	0.283
$4 \times 10^{6} < t_s < 2 \times 10^{8}$	53.2	0.335

A representation of the same type has been proposed by Johnston [13, 4] for his experiments relating to fast ^{239}Pu fission.

(b) The most widely used method consists in solving the coupled (differential) equations describing the time-dependent production and burnout of fission products, which need as input data the individual properties of the fission products: yields, decay schemes, half-lives, capture cross-sections. The number of data involved is extremely large, especially if one is interested in the emission spectra as well as in the residual power, although knowledge of the total energies emitted (E_β and E_γ) is in any case derived from the spectra, for part of the total energy released through decay is emitted in the form of antineutrinos. There is a shortage or a complete lack of data on short-lived fission products and, although often not specialists in such questions, the authors had to derive energy distribution rules or half-lives of nuclides with unknown decay schemes from nuclides with known decay schemes.

For the evaluations presented below the authors proceeded as follows:

(a) Fission yields were taken from the last compilation of Meek and Rider [15] for the evaluations of Tasaka, Devillers and Sporrer. For the evaluation of Tobias they were taken from Flynn and Glendenin [16] and Crouch [17]. For the evaluation of Battat et al. they were taken from Burris and Dillon [18], Weaver et al. [19] and Anderson [20];

(b) The Q-values relating to non-measured nuclides were those given by the formula by Myers and Swiatecki [21] for the evaluation of Tasaka and those of Garvey et al. [22] for the evaluation of Tobias and Devillers. The Q-values for measured nuclides were generally taken from the tables of Wapstra and Gove [23];

(c) For determining the energy partition among ß, γ and neutrino emission, Devillers, Tobias and Tasaka use the Fermi model to calculate the mean ß energy, subtracting from it the γ energy (obtained from the Q-value of the reaction) and, when the spectrum is not known, take $\frac{E_\beta + E_\gamma}{Q}$ = 0.58 (Tasaka), = 0.66 (Tobias) and as a function of the element (Devillers).

The decay schemes of the known nuclides are taken from Lederer's table of isotopes [24] and from Nuclear Data Sheets and rearranged in special lists: Tobias [25] and Devillers and de Tourreil [26], the list of the last-mentioned authors having the advantage of being presented in ENDF format.

The number of fission products taken into account varies from one list to another - about 600 in the case of Devillers and Tobias, 560 in the case of Tasaka et al. (125 stable nuclides), 243 in the case of Vossebrecker and 200 in the case of Battat et al. The names of the codes are as follows: FISP (Tobias), RASPA (Vossebrecker), Pepin (Devillers), FPIC (Battat and Dudziack), FPS (Tasaka and Sasamoto) and CINDER (England and Sporrer).

2.3. Results of the comparison

The results of the different evaluations and the ratios of these evaluations to the evaluation of Devillers (taken as reference for reasons of convenience) are presented in Tables I, II, III, IV and V and in Figs I, II, III, IV and V.

2.3.1. ^{235}U th

The results relating to ^{235}U th are naturally the most numerous.

For elemental fission (Table I,1; Fig. I) the most recent evaluations (Devillers, Tobias) are consistent to within 10% over the entire time range considered (1 sec to 10^8 sec). The evaluation of Tasaka et al. converges

with those of Devillers and Tobias at about 50 sec but diverges from them appreciably beyond 10^7 sec; this is almost certainly due to differences between the decay schemes of some long-lived nuclides (^{144}Pr, ^{90}Y, ^{90}Sr, $^{137}Ba = Cs$).

The evaluation of Vossebrecker converges with the reference evaluation at 10^3 sec, but the list used does not contain short-lived nuclides. The appreciable divergence beyond 10^7 sec is certainly due to an error, for it does not recur in the case of irradiation over a period of one year.

The evaluations of Battat et al. and Perkins agree to within some 5% with the reference evaluation between 10^3 sec and 10^8 sec.

The evaluation of England, derived from the C 17 case in his thesis (irradiation for 40 sec in a thermal flux of 10^{12}) is considerably higher (~ 35%) than the different evaluations for cooling times between 10^4 sec and 10^8 sec. We shall revert to this later.

The evaluation of Shure fluctuates in a regular manner around the mean, the deviation reaching 40% with a cooling time of 10 sec and then varying between +20% and -20%.

The oscillations obviously disappear if the elemental fission curve is integrated to obtain irradiation from one year at zero flux (Table I.2; Fig. I). The results are fewer but, in the light of the elemental fission curves just examined, the evaluations of Devillers, Tobias and Tasaka may be considered to agree to within 5% for all cooling times (as indicated above, Tasaka's evaluation diverges beyond 10^7 sec). The evaluations of Vossebrecker, Perkins and Battat converge with those of the authors just mentioned at 10^3 sec (leaving aside short-lived nuclides). Shure's evaluation is remarkably close to the recent evaluations for cooling times between 1 sec and 10^7 sec, but beyond 10^7 sec the divergence noted in the case of the elemental fission curves recurs (for $T_{ref} = 2 \times 10^8$ sec of cooling time $^{Shure}/_{Devillers} = 1.79$) without any clear cause, for in his evaluation Shure used the calculations of Perkins for long cooling times.

The results of England are appreciably higher than the mean for cooling times between 10 sec and 10^6 sec. Admittedly, the circumstances (irradiation for 10^4 hours at a constant flux of 2×10^{13}; results given in MeV per fission following irradiation) differ from those of the reference study, but this does not explain the divergence observed for the cooling times in question; Tobias makes the same comment in his critical study of experimental results [7]. The explanation for the difference has been given by Shure [27], who detected a slight error in the CINDER programme and gave the corrected result.

The results of England in Table I.1 were taken from his thesis. In

Table I.2, we present the results published by England in his thesis and also the two sets of values obtained by Shure after correcting the CINDER programme and England's data. It can be seen that, after the corrections have been made, the results agree closely with the other evaluations.

2.3.2. ^{239}Pu th, ^{233}U th, ^{235}U fast (Tables II, III and IV; Figs II, III and IV

There are far fewer evaluations to compare in this case, only Devillers and Tasaka having replied. The earlier remarks still apply: good agreement among the evaluations (to within 5%) up to 10^7 sec and then divergence due to long-lived products.

2.3.3. ^{239}Pu fast (Table V; Fig. V)

Here again there is little material on which to base a comparison. Tasaka's evaluation agrees to within 10% with the reference evaluation up to 10^7 sec, beyond which it diverges appreciably (at 10^8 sec, $^{\text{Tasaka}}$/Devillers = 1.7).

The evaluation of Battat and Dudziack converges with the reference evaluation before 10^3 sec and then remains in agreement with it to within 10%. Vossebrecker's evaluation behaves in the same way. Sporrer's [9], which - like England's - is based on the CINDER code, is appreciably higher than the reference evaluation; this does not seem to be due to the irradiation conditions (irradiation for 10^4 hours in a fast neutron flux of the order of 4×10^{15}).

2.4. Comments

The over-all impression gained from examining the evaluations is that there is good general consistency. The most recent evaluations (Devillers, Tobias) are in agreement for all cooling times. The older evaluations, which take into account a smaller number of fission products, agree with these for cooling times between 100 sec and 1000 sec. Beyond 10^7 sec there occur divergences which should be eliminated by examining the fission product decay schemes, which have in principle been measured accurately. Certain evaluations (England, Sporrer) are appreciably higher than the mean.

Before going on to examine the experimental results, let us consider what credence may be placed in the evaluations, bearing in mind the reliability of the basic data.

Fig. XI shows, for a ^{235}U thermal fission event and for an irradiation period of one year, the fraction of the residual power due to fission products for which the ß-spectra have been measured.

The unknown fraction (80% for 1 sec, 40% for 100 sec, <1% for 1000 sec) is attributable to nuclides whose ß-spectra have not been measured and for which the Q_β-value is generally derived from mass formulas whose precision is of the order of 20%. For these nuclides we have been obliged to make assumptions

about the Q_β-distribution, which introduces an additional uncertainty. Moreover, detailed examination of the decay chains shows that a certain number of short-lived nuclides (<1 min) have not been identified and that for those which have been identified the uncertainty as regards their half-lives is often considerable, the general tendency being to underestimate.

Accordingly, one should not expect a precise evaluation of residual powers for cooling times <1000 sec, the agreement among evaluations being due to the fact that the sources of data are very often the same. Only comparison with experiment will indicate the error which may be expected. Beyond 10^3 sec, for nuclides which are "known" in the sense intended in Fig. XI, the Q_β-value has in most cases been determined experimentally and compiled by Wapstra with a mean precision of 2.5%.

There remain the problems of ß, γ, $\bar{\nu}$ (antineutrino) partition, for many ß-spectra are still imprecise. For example, in the case of nuclides accounting for about 10% after a cooling time of 2000 sec (^{94}Y, ^{138}Cs) it is safe to say that the ß-spectra under consideration do not agree to within 10% with the Q_β-value and the γ-spectra recently measured by means of a Ge(Li) detector:

$$Q_{\beta,\text{measured}} \neq \Sigma E_{\beta,\max} I_\beta + \Sigma E_\gamma I_\gamma$$

The expected error due to the ß, γ distribution of a nuclide with known Q_β should not, however, exceed ± 5% of the Q_β-value in the case of these imprecise ß-spectra. Consistency is achieved only very gradually, and for ^{140}La, which accounts for more than 50% after a cooling time of 8×10^6 sec,

$$Q_{\beta,\text{measured}} = 3.767 \text{ MeV (Wapstra)}$$

$$(\Sigma E_\beta I_\beta + \Sigma E_\gamma I_\gamma = 3.715 \text{ MeV} - \text{i.e. a difference of } 1.4\%).$$

For long cooling times, the precision of the evaluation should then be of the same order of magnitude as the precision of the Q_β-value. However, because of the very large number of parameters involved, the value is generally very difficult to give without making a numerical study, which would involve lists containing – in addition to data – the associated error bars. No such lists exist yet, although attempts are being made to compile some.

Let us now consider the agreement between evaluations and experimental results.

3. COMPARISON OF EVALUATIONS AND EXPERIMENTAL RESULTS

Although, as we have seen, quite a large number of residual power evaluations have been carried out (those mentioned in section 2 are by no means all), there have been very few experiments, and these have concerned ß and γ emissions separately rather than the total power emitted. As in the case of the evalua-

tions, we have confined ourselves to comparing the total energies emitted, whether ß or γ, leaving aside the spectra, which do not lend themselves so readily to numerical comparisons.

The experimental results given below are generally the results recorded by the experimenters themselves, except where they have made a point of providing the spectrum, in which case integration (sometimes on the basis of figures) has been performed in order to obtain $E_ß$ or $E_γ$. The Figures corresponding to the Tables show the ratio of the experimental value to the corresponding calculated value (Devillers) and, wherever possible, enable one to compare (still in terms of ratios) different evaluations with that of Devillers.

A comparison of this type has been made by Costa and de Tourreil [28] and later by Tobias [29] using the same data, and the material presented here is based largely on these two comparisons.

3.1. ^{235}U th

This is obviously the reaction for which the most measurements have been performed.

3.1.1. ^{235}U th $E_ß$ (Tables VI.1, 2, 3, 4, 5, 6, 7, 8; Figs VI.1, 2)

A. McNair et al. (1968) [30] measured the ß energy emitted by the fission products of ^{235}U th after irradiations at a constant fission rate for 10^2, 10^3, 10^4 and 10^5 sec by means of an NE-102 ß-scintillator; the fission rate during irradiation was measured in relation to gold.

The stated precision was ± 4%, the semi-experimental gamma correction lay between 5% and 20% and the stated over-all precision was of the order of ± 4%.

T.D. McMahon et al. (1970) [31] performed the same experiments, but measured the gamma contribution by means of a magnetic deflector. The fission rate was measured in relation to fission chambers or standards (^{99}Mo, ^{140}Ba-La). The stated precision was also ± 4%, but the deviation from McNair's results was as high as 30% - especially in the case of long cooling times.

J.W. Kutcher and M.E. Wymann (1966) [32] measured the ß-spectrum following irradiations for one hour and three hours and after a neutron pulse. Unfortunately, they did not give any table of values and the results presented in Table VI contain the error involved in reading the curves and performing the integration. The ß energies were measured by means of a plastic scintillator. The fission rate was measured by means of a fission chamber, the γ energies being eliminated by the coincidence method. The stated over-all precision was ± 7.2%; after integration it was certainly up to ± 10%.

Tsoulfanidis et al. (1970) [33] repeated the preceding spectrum

measurements, after a neutron pulse and irradiation for 30 000 sec, and gave the total energy. The fission rate was measured by means of a fission chamber and the γ energies separated out. These authors gave values for total ß energy, the stated precision being of the order of ± 10%. Their results are in close agreement with those of Kutcher and Wymann.

Comments

The residual ß power evaluations of Devillers, Tobias, Scobie and Scott [34] and Battat et al. (for cooling times greater than 10^3 sec) agree to within ± 5% as in the case of total residual power. The disagreement among the experimental results is quite a lot in excess of the errors stated by experimenters, being at least ± 10%: McNair/McMahon = 1.3 at 10^4 sec.

Under these conditions it is difficult to say whether the theoretical evaluations are at fault. It would seem (if one leaves aside McMahon's experiments) that for one fission event the theoretical evaluations underestimate the residual power values for very short cooling times (<10 sec) by a factor of < 1.5, that between 10 sec and 10^3 sec they underestimate the true residual power by about 20% and that from 10^3 sec to about 10^5 sec the theoretical estimate is correct.

3.1.2. ^{235}U th E_γ (Table VII; Fig. VII)

It is more difficult to measure γ energy directly than ß energy. The measurements were made longer ago and the inconsistency of the experimental results is greater.

Maienschein et al. (1958) [35] measured the γ-spectrum following a fission event by means of a sodium iodide spectrometer after cooling times of 0–1500 sec. The results are given in the form of curves, which have to be integrated in order to obtain the total energy. The authors put the precision at ± 15%.

Bunney and Sam (1969) [36], also using a sodium iodide spectrometer (total absorption), extended the preceding experiment from 15 minutes to three hours after the fission event. The results are presented in tabular form and the precision stated by the experimenters is better than 10%, which is the stated error in the fission rate.

Sakharov et al. (1957) [37] performed measurements with cooling times similar to those involved in those of Bunney and Sam; the published results are very similar.

Petrov (1960) [38] performed measurements of absorbed energy using ionization chambers. However, the results published by him are not really compatible with the results of Sakharov et al. or with the calculated values.

Comments

For cooling times greater than 10^3 sec (10^3–10^6 sec) the theoretical evaluations are lower than the measured values, which agree among themselves to within a factor of ~ 1.2. For shorter times, the deviation is as great as 40% (for a cooling time of 7 sec) relative to the evaluation of Devillers and about 25% relative to the evaluation of Tobias.

3.1.3. ^{235}U th $E_\beta + E_\gamma$ (Table VIII; Fig. VIII)

Lott et al. (1973) [39] used a calorimetric method to measure the total energy following a fission event. The stated precision is of the order of ± 5%. The fission rate was measured relative to ^{140}Ba-La. The cooling times lay between 70 sec and 1.5 x 10^7 sec. Beyond 10^3 sec the experiment/calculation ratio is close to 1.05, which means that the calculated values have more or less returned to within the error limits of the experiment. Below 5 x 10^2 sec (70 sec < t < 500 sec) the calculated values exceed the experimental values by a factor of as much as 1.3.

3.2. ^{239}Pu th E_β (Table IX; Fig. IX)

McNair et al. [40] carried out with plutonium virtually the same series of measurements as with ^{235}U. The agreement between experiment and evaluation is considerably better than for ^{235}U, the experiment/evaluation ratio being of the order of ± 1.1 for short cooling times and tending towards unity for cooling times of 10^4–10^5 sec.

^{239}Pu th E_γ (Table X)

We have only the experiment of Petrov [38], which produced the same figures as for ^{235}U th. The experimental results are considerably higher than the evaluation.

3.3. ^{233}U th E_β (Table XI)

Scobie and Scott (1970) [41] measured, following irradiation for 10^4 sec, the ratio of the emission by ^{235}U to that by ^{233}U using the same equipment as that employed by McMahon. The calculated results presented by Scobie and Scott agree to within 1% with the calculated result of Devillers, but the divergence between calculation and experiment may reach 15% for a cooling time of 100 sec, tending towards zero for cooling times greater than 10^4 sec. Using McMahon's experiment as a reference, the ratio $\frac{E_{\beta,\text{exp.}}\,^{233}U}{E_{\beta,\text{theo.}}\,^{233}U}$ for a cooling time of 100 sec is found to be of the order of 0.87.

3.4. ^{235}U fast E_γ (Table XII.1, 2)

Bunney and Sam (1967) [42] used the technique described above for

^{235}U fast and ^{238}U fast; the results are given in the same form as before. The experiment/calculation ratio is of the order of 1.5 for all cooling times (10^3–10^5 sec).

Fisher and Engle (1964) [43] measured the γ-spectra resulting from the fast fission of ^{233}U, ^{235}U, ^{238}U, ^{232}Th and ^{239}Pu after cooling times of less than 35 sec. The experiment/Devillers ratio is of the order of 1.6 for ^{235}U and 2 for ^{239}Pu. The evaluation of Tobias is considerably closer to the experimental values.

3.5. ^{239}Pu fast $E_\beta + E_\gamma$ (Table XIII.1)

Johnston (1965) [13], using a calorimetric technique and with cooling times of 40–150 days, measured the total energy emitted by the fission products of a plutonium sample irradiated at Dounreay. The experiment/calculation (Tobias) ratio is of the order of 1.05.

^{239}Pu fast + ^{235}U fast (Table XIII.2)

Lott et al. (1973) [44] measured, using a calorimetric technique and with cooling times of 300–1600 days, the residual power of fuel elements irradiated for 172–1212 days in the Rapsodie reactor. With the exception of one measurement involving major corrections, the experiment/calculation ratio lay between 1.01 and 1.04.

3.6. General comments on the experiments

The general impression gained from an examination of the experimental results as a whole is rather bad. However, we have seen above that it was very difficult to assign an error to the evaluations, whereas comparison of calculation with experiments enables one to do so.

The over-all experiment/evaluation ratio lies in the ranges:

0.9–2.2 for γ measurements;

0.7–1.5 for β measurements;

0.83–1.08 for β + γ measurements.

The range is greatest in the case of γ measurements; it includes Petrov's measurements, the results of which (as we have already pointed out) are incompatible with the other results, and the measurements of Bunney and Sam, which were performed under irradiation conditions unlike those obtaining in a fast reactor, so that problems of fission yield arise. The table above does not change if one ignores these two experiments, but the extreme appears only in the case of short cooling times.

The following table shows the observed deviations as a function of cooling time:

Sec	< 10	10–100	100–1000	1000–10^5	> 10^5	No. of experiments
ß	0.8–1.76	0.75–1.30	0.75–1.1	0.66–1.1	0.96–1	4
γ	1.1–2.6	0.95–1.56	1.12–1.25	1.15–1.25	0.95–1.25	4
ß + γ		0.83–0.85	0.85–1.05	1.16–1.06	1.02–1.11	3

This table gives an idea of the error to be expected if one carries out an evaluation. It is fairly obvious that below 10 sec evaluations are lower than experimental values by a factor of 1.5–2; with longer cooling times the extreme upper and lower values come closer together and the mean value approaches unity (it reaches unity only if $t > 10^5$ sec, but only very few experiments have been performed with such long cooling times).

4. CONCLUSIONS AND RECOMMENDATIONS

In the first part of this paper, where we compared evaluations, our conclusions were rather optimistic: the evaluations were consistent to within a few per cent. From an examination of the second part it appears that the evaluations are not consistent with the experimental values (although the deviations do not exceed a factor of 2) and that the experimental values do not agree among themselves.

That being so, what recommendations can one make with a view to improving the situation?

As regards evaluations, it is essential that authors make the effort to indicate the precision of their results in direct relation to the precision of the basic data, for nothing is gained if evaluations involve systematic errors (neglected nuclides) and if the precision is not indicated numerically.

As regards the basic data, there are many short-lived nuclides about which little is known. Efforts should be concentrated on measuring β decay spectra, which have so far been studied far less than γ-spectra.

As regards experiments, the role of an integral experiment should be to throw more light on the precision of an evaluation and to replace it if the evaluation is imprecise (short cooling times).

The calorimetric technique, which is rarely used as it does not lend itself to measurements following short cooling times (<100 sec), is without doubt the most simple and precise method. If it gives poor results, separate ß and γ measurements should be performed.

It would be very interesting to measure ß, γ and ß + γ following irradiation under similar conditions and cooling times which permitted the use of all techniques, as this would indicate the internal consistency of experiments and enable experimenters to qualify their results by intercomparison.

It is certainly not worth carrying out many experiments with a large number of different nuclides; a good series of experiments with ^{235}U th would be more valuable than a large number of experiments involving all fissionable nuclides.

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TABLE I.1:
Thermal fission of ^{235}U; 1 fission; $E_\beta + E_\gamma$ [MeV s^{-1} fission $^{-1}$]

TCOOL (s)	DEVILLERS	TASAKA	TOBIAS	VOSSE-BRECKER	BATTAT DUDZIACK	PERKINS	SHURE	ENGLAND
1 0	.651 0		.722 0	.198 -1			.77 0	
2	.445		.503	.194			.368	
5	.228		.249	.187			.139	
1 1	.128	.112 0	.130	.179	.291 -1		.182	
2	.678 -1	.576 -1	.677 -1	.164			.805 -1	
5	.276	.258	.292	.126			.272	
1 2	.133	.139	.144	.847 -2	.101 -1	.696 -2	.120	
2	.590 -2	.665 -2	.649 -2	.458	.560 -2		.821 -2	
5	.194	.215	.210	.168			.253	
1 3	.914 -3	.965 -3	.942 -3	.818 -3	.878 -3	.846 -3	.104	
2	.446	.463	.445	.413	.415 -3		.427 -3	
5	.144	.149	.144	.140			.132	
1 4	.579 -4	.594 -4	.592 -4	.563 -4	.584 -4	.619 -4	.542 -4	.672 -4
2	.230	.230	.233	.226	.226 -4		.222	
5	.699 -5	.723 -5	.705 -5	.710 -5			.687	.887 -5
1 5	.259	.274	.263	.265	.258 -5	.268 -5	.282	.381 -5
2	.926 -6	.981 -6	.925	.941 -6	.950 -6		.116	
5	.307	.321	.311 -6	.311			.358 -6	
1 6	.157	.164	.162	.162 -6	.166 -6	.157 -6	.147	.202 -6
2	.742 -7	.776 -7	.764 -7	.774 -7	.788 -7		.604 -7	
5	.229	.235	.235	.237			.203	
1 7	.965 -8	.986 -8	.971 -8	.102	.961 -8	.964 -8	.805 -8	.142 -7
2	.359	.370	.354	.421 -8	.357 -8		.319	
5	.697 -9	.759 -9	.689 -9	.137			.939 -9	
1 8	.225 -9	.257	.226	.903 -9	.228 -9	.231 -9	.372	
2	.857 -10	.104		.767			.147	
5	.589 -10	.719 -10		.743				
1 9	.400							

TABLE I.2:
Thermal fission of ^{235}U; irradiated for 1 year at 1 fission/sec.;
$E_\beta + E_\gamma$ /MeV/fission/

TCOOL (S)	DEVILLERS	TASAKA	VOSSE-BRECKER	SHURE	ENGLAND 104 hours (Thesis)	ENGLAND corrected by SHURE	ENGLAND modified data
1 0	0.115 2		.675 1	.118 2			
2	.109		.673	.113			
5	.100		.667	.107			
1 1	.917 1	.916 1	.658	.993 1	.982 1	.967 1	.931 1
2	.825	.838	.641	.874			
5	.700	.727	.598	.738			
1 2	.605	.634	.546	.649	.716	.699	.664
2	.518	.543	.483	.563			
5	.420	.436	.403	.430			
1 3	.356	.367	.346	.351	.438	.414	.394
2	.293	.303	.288	.285			
5	.218	.225	.218	.216			
1 4	.173	.179	.174	.174	.220	.192	.185
2	.137	.143	.139	.140			
5	.100	.105	.102	.104			
1 5	.797 0	.833 0	.808 0	.832 0	.113	.940 0	.872 0
2	.644	.671	.652	.655			
5	.496	.516	.502	.468			
1 6	.389	.403	.393	.354	.545 0	.45	.436
2	.284	.293	.285	.265			
5	.165	.170	.164	.147			
1 7	.990 -1	.102	.975 -1	.911 -1	.157	.118	.116
2	.470	.496 -1	.464	.517			
5	.150	.164	.146	.212			
1 8	.559 -2	.645 -2	.550 -2	.975 -2			
2	.256	.311	.264	.421 -2			
5	.183	.223	.181				
1 9	.124						

	1 fission [MeV S^{-1}]			Irradiation for 1 year [MeV / fission]	
TCOOL(S)	DEVILLERS	TASAKA		DEVILLERS	TASAKA
1 0	.426 0			.959 1	
2	.288			.924	
5	.152			.863	
1 1	.922 - 1	.785 - 1		.805	.799 - 1
2	.530	.440		.735	.742
5	.235	.211		.632	.653
1 2	.116	.117		.550	.576
2	.525 - 2	.567 - 2		.473	.494
5	.181	.195		.385	.402
1 3	.881 - 3	.933 - 3		.323	.338
2	.423	.448		.263	.276
5	.129	.136		.193	.204
1 4	.485 - 4	.508 - 4		.154	.163
2	.181	.185		.125	.132
5	.590 - 5	.600 - 5		.958 0	.103
1 5	.241	.250		.771	.834 0
2	.944 - 6	.100		.623	.678
5	.324	.343 - 6		.467	.513
1 6	.154	.164		.357	.398
2	.679 - 7	.727 - 7		.258	.292
5	.201	.218		.153	.181
1 7	.841 - 8	.952 - 8		.972 - 1	.121
2	.346	.430		.539	.725 - 1
5	.961 - 9	.138		.212	.310
1 8	.315	.468 - 9		.749	.111
2	.756 - 10	.107		.213	.297
5	.386	.512 - 10		.120	.160
1 9	.262			.818 - 2	

TABLE II :

Thermal fission of ^{239}Pu : $E_\beta + E_\gamma$

	1 fission [MeV S^{-1}]		Irradiation of 1 year [MeV / fission]	
T_{cool} (S)	DEVILLERS	TASAKA	DEVILLERS	TASAKA
1 0	.319 0		.965 1	
2	.234		.938	
5	.236		.885	
1 1	.849 -1	.701 -1	.832	.783 1
2	.494	.401	.768	.731
5	.225	.196	.671	.650
1 2	.115	.108	.591	.581
2	.536 -2	.529 -2	.513	.511
5	.183	.185	.423	.421
1 3	.876 -3	.881 -3	.361	.361
2	.435	.436	.300	.301
5	.146	.148	.226	.227
1 4	.610 -4	.614 -4	.179	.180
2	.250	.245	.141	.141
5	.722 -5	.719 -5	.101	.102
1 5	.254	.261	.805 0	.809 0
2	.918 -6	.956 -6	.656	.654
5	.320	.323	.505	.499
1 6	.163	.164	.393	.386
2	.764 -7	.759 -7	.283	.278
5	.232	.225	.162	.159
1 7	.957 -8	.923 -8	.947 -1	.941 -1
2	.342	.341	.432	.445
5	.618 -9	.665 -9	.133	.145
1 8	.211	.238	.538 -2	.612 -2
2	.918 -10	.109	.277	.329
5	.646	.773 -10	.201	.241
1 9	.437		.136	

TABLE III: Thermal fission of ^{233}U : $E_\beta + E_\gamma$

TABLE IV:
Fast fission of ^{235}U; $E_\beta + E_\gamma$

	1 fission MeV s^{-1}			one year irradiation MeV / fission		
TCOOL (S)	DEVILLERS	TASAKA	VOSSE-BRECKER	DEVILLERS	TASAKA	VOSSE-BRECKER
1 0	.584 0		.155 -1	.112 2		.560 1
2	.405		.153	.107		.558
5	.213		.147	.985 1		.557
1 1	.122	.110 0	.140	.905	.925 1	.546
2	.661 -1	.585 -1	.127	.816	.847	.533
5	.272	.265	.969 -2	.693	.732	.500
1 2	.131	.142	.652	.599	.636	.460
2	.580 -2	.671 -2	.360	.513	.541	.411
5	.191	.214	.142	.417	.436	.346
1 3	.911 -3	.973 -3	.740 -3	.353	.367	.295
2	.445	.469	.372	.290	.302	.244
5	.142	.150	.117	.215	.225	.181
1 4	.569 -4	.592 -4	.438 -4	.171	.178	.146
2	.226	.228	.172	.136	.142	.119
5	.686 -5	.712 -5	.577 -5	.999 0	.105	.904 0
1 5	.255	.270	.227	.792	.330 0	.724
2	.929 -6	.981 -6	.838 -6	.641	.670	.588
5	.312	.325	.275	.491	.513	.455
1 6	.156	.162	.140	.383	.400	.360
2	.726 -7	.755 -7	.663 -7	.279	.292	.267
5	.223	.232	.210	.164	.173	.163
1 7	.949 -8	.986 -8	.963 -8	.991 -1	.106	.104
2	.358	.381	.440	.482	.533 -1	.565 -1
5	.738 -9	.854 -9	.153	.158	.186	.212
1 8	.238	.288		.586 -2	.712 -2	.750 -2
2	.833 -10	.103		.247	.305	.214
5	.550	.677 -10		.171	.211	.120
1 9	.373			.116		

T_{cool} (s)	DEVILLERS	TASAKA	BATTAT DUDZIACK	VOSSE-BRECKER
1 0	.408 0			.155 - 1
2	.276			.153
5	.147			.147
1 1	.901 - 1	.781 - 1	.196 - 1	.140
2	.522	.447		.127
5	.233	.220		.969 - 2
1 2	.116	.119	.781 - 2	.652
2	.523 - 2	.573 - 2	.453	.360
5	.181	.199		.142
1 3	.888 - 3	.956 - 3	.848 - 3	.740 - 3
2	.426	.456	.417	.372
5	.129	.135		.117
1 4	.482 - 4	.503 - 4	.498 - 4	.438 - 4
2	.180	.184	.181	.172
5	.581 - 5	.593 - 5		.577 - 5
1 5	.238	.245	.235 - 5	.227
2	.937 - 6	.976 - 6	.915 - 6	.838 - 6
5	.324	.338		.275
1 6	.155	.161	.146	.140
2	.678 - 7	.707 - 7	.670 - 7	.663 - 7
5	.196	.213		.210
1 7	.816 - 8	.933 - 8	.897 - 8	.963 - 8
2	.341	.428	.382	.44
5	.979 - 9	.143 - 9		.153
1 8	.324	.486	.351 - 9	
2	.778 - 10	.110 - 10		
5	.389	.518		
1 9	.264		.271 - 10	

TABLE V. 1 :

Fast fission of ^{239}Pu: $E_\beta + E_\gamma$ [MeV s^{-1}]

T_{cool} (S)	DEVILLERS	TASAKA	SPORRER 10^{-4} Pu	VOSSE-BRECKER
1 0	. 95 1			. 56 1
2	.916			. 56
5	.858			. 55
1 1	.801	.807 1	.103 2	.547
2	.733	.749	.100	.533
5	.631	.658	.86 1	.500
1 2	.55	.579	.67	.460
2	.473	.496	.56	.411
5	.384	.402	.51	.346
1 3	.322	.338	.47	.295
2	.261	.274	.39	.243
5	.192	.201	.31	.181
1 4	.153	.160	.261	.146
2	.124	.130	.21	.119
5	.949 0	.100	.16	.904 0
1 5	.765	.817 0	.126	.724
2	.618	.665	.100	.59
5	.463	.503	.76 0	.455
1 6	.353	.390	.579	.360
2	.253	.287	.44	.267
5	.150	.179	.24	.163
1 7	.958 -1	.121	.164	.104
2	.540	.737 -1		.565 -1
5	.216	.320		.212
1 8	.772 -2	.115		.750
2	.219	.305 -2		.214
5	.121	.167		.120

TABLE V.2: Fast fission of ^{239}Pu: $E_\beta + E_\gamma$ [MeV/fission], irradiation for 1 year

	EXPERIMENTS		
TCOOL (S)	DEVILLERS	Mc NAIR	Mc MAHON
1 0	.140 1	.209 1	
2	.116	.148	
3.5	.927 0		
4	.872		
5	.782	.926 0	
7	.649		
1 1	.519	.565	.512 0
1.5	.389		.353
2	.310	.288	.268
3	.219		.180
5	.138	.108	.105
7	.991 - 1		.725 - 1
1 2	.677	.507 - 1	.481
1.5	.421		.292
2	.295	.223	.206
3	.177		.128
5	.952 - 2	.820 - 2	.719 - 2
7	.650		.509
1 3	.442	.420	.366
1.5	.285		.236
2	.204		.169
3	.121		.977 - 3
5	.593 - 3		.457
7	.378		.285
1 4	.244		.183
1.5	.155		.116
2	.113		
3	.711 - 4		
5	.367		
7	.219		

TABLE VI. 1 :

Thermal fission of ^{235}U - $E\beta$, [MeV/fission] Tir = 10 s

	EXPERIMENTS		
TCOOL (S)	DEVILLERS	Mc NAIR	Mc MAHON
1 0	.307 1	.354 1	
2	.277	.293	
3.5	.246		
4	.238	.218	.229 1
5	.225		.208
7	.203		.181
1 1	.180	.160	.155
1.5	.153		.126
2	.134	.109	.104
3	.109		.850 0
5	.794 0	.605 0	.582
7	.621		.454
1 2	.461	.341	.328
1.5	.314		.219
2	.233	.173	.168
3	.151		.113
5	.866 - 1	.729 - 1	.645 - 1
7	.609		.491
1 3	.422	.382	.350
1.5	.275		.250
2	.198	.182	.165
3	.118		.967 - 2
5	.586 - 2	.530 - 2	.454
7	.375		.285
1 4	.243		.178
1.5	.154		.111
2	.112		.815 - 3
3	.710 - 3		.538
5	.366		.298
7	.219		.179

TABLE VI.2 : Thermal fission of ^{235}U; E_β [MeV/fission]; Tir = 10^2s

	EXPERIMENTS		
TCOOL (S)	DEVILLERS	Mc NAIR	Mc MAHON
1 0	.434 1	.445 1	
3.5	.371		.347 1
5	.349	.314	.295
7	.326		.272
10 1	.301	.255	.241
1.5	.271		.211
2	.250	.200	.195
3	.219		.167
5	.181	.141	.136
7	.157		.118
1 2	.132	.103	.979 0
1.5	.106		.795
2	.894 0	.722 0	.685
3	.695		.540
5	.496	.430	.404
7	.391		.323
1 3	.298	.265	.247
1.5	.209		.174
2	.156	.140	.126
3	.788 - 1		.776 -1
5	.525	.463 - 1	.394
7	.348		.252
1 4	.232	.213	.166
1.5	.149		.111
2	.110		.820 -2
3	.696 - 2		.528
5	.361		.287
7	.216		.184
1 5	.121		.118
1.5	.615 - 3		

TABLE VI.3 : Thermal fission of ^{235}U : E_β [MeV/fission]; $T_{ir} = 10^3$ s.

TCOOL (S)	CALCULATIONS		EXPERIMENTS	
	DEVILLERS	SCOBIE	MAC NAIR	MAC MAHON
1 0	. 516 1		. 524 1	
2	. 484		. 464	
5	. 429		. 388	. 362 1
7	. 407			. 332
1 1	. 381		. 328	. 308
1.5	. 351			. 277
2	. 330		. 273	. 257
3	. 299			. 228
5	. 260		. 213	. 198
7	. 234			. 178
1 2	. 208	. 209 1	. 173	. 158
1.5	. 181			. 139
2	. 163	. 167	. 140	. 126
3	. 139			. 111
5	. 119	. 117	. 102	. 905 0
7	. 985 0			. 790
1 3	. 830	. 859 0	. 747 0	. 662
1.5	. 664			. 508
2	. 554	. 571	. 492	. 417
3	. 416			. 307
5	. 282		. 250	. 195
7	. 216			. 149
1 4	. 162	. 164	. 149	. 110
1.5	. 118			. 776 - 1
2	. 890 - 1	. 885 - 1	. 84 -1	. 589
3	. 588			. 377
5	. 318			. 220
7	. 196			. 141
1 5	. 112	. 104	. 117	. 806
1.5	. 586 - 2			

TABLE VI.4 : Thermal fission of ^{235}U E Tir = 10^{4}s [MeV/fission]

	CALCULATIONS			EXPERIMENTS	
TCOOL (S)	DEVILLERS	SCOBIE	TOBIAS	Mc.NAIR	Mc.MAHON
1 0	.561 1		.566 1	.569 1	
2			.533	.509	
5	.475		.475	.432	.393 1
1 1	.427		.425	.372	.339
1.5	.397				.308
2	.375		.374	.317	.288
3	.344				.259
5	.305		.306	.256	.229
7	.280				.209
1 2	.254	.254 1	.255	.216	.189
1.5	.226				.170
2	.208	.212	.209	.183	.157
3	.185				.142
5	.158	.161	.158	.144	.121
7	.143				.109
1 3	.127	.129	.125	.116	.959 0
1.5	.109				.798
2	.970 0	.918 0	.956 0	.897 0	.700
3	.815				.578
5	.649		.641	.608	.445
7	.557				.380
1 4	.469	.462	.463	.449	.319
1.5	.377				.258
2	.316	.307	.313	.313	.216
3	.236				.163
5	.149		.148		
7	.105				
1 5	.674 -1	.613 - 1	.674 - 1	.703 - 1	
1.5	.402				
2	.275	.242	.272	.264	
5	.979 -2		.97	.970	

TABLE VI.5 : Thermal fission of ^{235}U, Tir = 10^5, MeV/fission

TABLE VI.6 : ^{235}U th, E_β
1 fission, Tir = 0
(TSOULFANIDIS)

Δ TCOOL (S)	DEVILLERS MeV S^{-1}	TSOULFANIDIS
10-16	.584.- 1	.777
60-72	.114 - 1	.106 - 1
180-228	.299 - 2	.252 - 2
900-1020	.465 - 3	.466 - 3
3600-3900	.890 - 4	.980 - 4

Δ TCOOL (S)	DEVILLERS MeV/fission	TSOULFANIDIS
0-12	.456 - 1	.498 - 1
15-27	.352	.351
60-72	.264	.258
180-240	.184	.187
900-1020	.108	.109
3600-3900	.550 0	.518 0
10800-11100	.292	.267

TABLE VI.7 : ^{235}U th, E_β
Irradiation for 29700 s
(TSOULFANIDIS)

Δ TCOOL (S)	DEVILLERS MeV S^{-1}	KUTCHER WYMAN
0 - 5	.550 0	.836
5 - 10	.963 - 1	.170
10 - 15	.603 - 1	.800 - 1
15 - 25	.390	.445
60 - 70	.116	.109
180 - 225	.302 - 2	.247 - 2
900 - 1020	.465 - 3	.423 - 3
3600 - 3900	.89 0 - 4	.990 - 4
7200 - 7500	.355 - 4	.491 - 4

TABLE VI.8 : ^{235}U th, E_β
1 fission, Tir = 0
(KUTCHER + WYMAN)

TABLE VII:
Thermal fission of ^{235}U; Tir = 0, 1 fission, E_γ [MeV s^{-1} fission $^{-1}$]

	CALCULATIONS			EXPERIMENTS			
TCOOL (S)	DEVILLERS	TOBIAS	BATTAT	MAIENSCHEIN	BUNNEY SAM	SAKHAROV	PETROV
1 0	.272 0	.330 0		.315 0			.369 0
2	.185	.226		.254			.261
5	.970 -1	.109		.138			.136
1 1	.552	.567 -1	.736 -2	.616 -1			.739 -1
2	.297	.308		.283			.445
5	.125	.142		.151			.190
1 2	.613 -2	.738 -2	.493 -2	.770 -2			.909 -2
2	.286	.337	.294	.323			.439
5	.978 -3	.108		.110			.168
1 3	.470	.491 -3	.447 -3	.593 -3	.630 -3		.810 -3
2	.242	.241	.212	.372 (1500 s)	.325		.390
5	.846 -4	.833 -4			.990 -4	.100 -3	.150
1 4	.335	.342	.316 -4		.380	.396 -4	.720 -4
2	.118	.121	.111 -4		.148	.144	.250
5	.333 -5	.336 -5			.420 -5	.422 -5	
1 5	.138	.138	.139 -5		.170	.170	
2	.546 -6	.543 -6	.564 -6		.675 -6	.697 -6	
5	.201	.200				.249	
1 6	.103	.103	.111			.100	
2	.465 -7	.461 -7	.509 -7				
5	.129	.127					
1 7	.509 -8	.498 -8	.501 -8				

TCOOL (S)	DEVILLERS	TOBIAS (Calc)	LOTT et al (1973)
7 1	0.01953	0.02101	0.01629
1 2	0.01328	0.01445	0.01127
1.5	8.339×10^{-3}	9.141×10^{-3}	7.347×10^{-3}
2	5.903×10^{-3}	6.493×10^{-3}	5.341×10^{-3}
3	3.583×10^{-3}	3.941×10^{-3}	3.300×10^{-3}
5	1.936×10^{-3}	2.096×10^{-3}	1.899×10^{-3}
7	1.328×10^{-3}	1.405×10^{-3}	1.365×10^{-3}
1 3	9.145×10^{-4}	9.416×10^{-4}	9.631×10^{-4}
1.5	6.060×10^{-4}	6.105×10^{-4}	6.454×10^{-4}
2	4.461×10^{-4}	4.450×10^{-4}	4.763×10^{-4}
3	2.774×10^{-4}	2.750×10^{-4}	2.908×10^{-4}
5	1.438×10^{-4}	1.439×10^{-4}	1.529×10^{-4}
7	9.232×10^{-5}	9.367×10^{-5}	9.806×10^{-5}
1 4	5.786×10^{-5}	5.934×10^{-5}	6.111×10^{-5}
1.5	3.379×10^{-5}	3.468×10^{-5}	3.464×10^{-5}
2	2.300×10^{-5}	2.346×10^{-5}	2.286×10^{-5}
3	1.358×10^{-5}	1.374×10^{-5}	1.355×10^{-5}
5	6.991×10^{-6}	7.090×10^{-6}	7.216×10^{-6}
7	4.365×10^{-6}	4.445×10^{-6}	4.416×10^{-6}
1 5	2.592×10^{-6}	2.645×10^{-6}	2.674×10^{-6}
1.5	1.418×10^{-6}	1.447×10^{-6}	1.493×10^{-6}
2	9.257×10^{-7}	9.454×10^{-7}	9.844×10^{-7}
3	5.343×10^{-7}	5.470×10^{-7}	5.732×10^{-7}
5	3.074×10^{-7}	3.162×10^{-7}	3.261×10^{-7}
7	2.230×10^{-7}	2.300×10^{-7}	2.424×10^{-7}
1 6	1.567×10^{-7}	1.617×10^{-7}	1.676×10^{-7}
1.5	1.022×10^{-7}	1.052×10^{-7}	1.094×10^{-7}
2	7.422×10^{-8}	7.633×10^{-8}	7.896×10^{-8}
3	4.552×10^{-8}	4.677×10^{-8}	4.838×10^{-8}
5	2.289×10^{-8}	2.344×10^{-8}	2.474×10^{-8}
7	1.476×10^{-8}	1.502×10^{-8}	1.60×10^{-8}
1 7	9.66×10^{-9}		1.043×10^{-8}
1.5	5.69×10^{-9}		6.19×10^{-9}

TABLE VIII : Thermal fission of ^{235}U, $E_\beta + E_\gamma$ [MeV S^{-1} $fission^{-1}$]

TOOL	(S)	Tir 10 s		Tir 10^2 s		Tir 10^3 s		Tir 10^4 s		Tir 10^5 s	
		DEVILLERS	MAC - NAIR	DEVILLERS	MAC-NAIR	DEVILLERS	MAC-NAIR	DEVILLERS	MAC-NAIR	DEVILLERS	MAC - NAIR
2	1	.243 0	.242 10^0	.111 10^1	.104 10^1	.215 10^1	.200 10^1	.284 10^1	.276 10^1	.318 10^1	.314 10^1
5		.115	.103	.676 10^0	.601 10^0	.160	.147	.228	.223	.262	.255
1	2	.581 - 1	.512 10^{-1}	.400	.351	.118	.111	.185	.183	.219	.217
2		.260	.229	.208	.185	.817 10^1	.794 10^0	.145	.148	.179	.181
5		.880 - 2	.870 10^{-2}	.803 10^{-1}	.796 10^{-1}	.460	.487	.101	.107	.134	.140
1	3	.411	.440	.391	.418	.272	.300	.722 10^0	.771 10^0	.105	.109
2				.178	.191	.139	.154	.466	.485	.777 10^0	.796 10^0
5				.492 10^{-2}	.490 10^{-2}	.436 10^{-1}	.457 10^{-1}	.221	.221	.496	.500
1	4					.176	.184	.120	.119	.352	.354
2								.638 10^{-1}	.624 10^{-1}	.241	.245
5								.245	.233	.123	.123
1	5									.611 10^{-1}	.552 10^{-1}

TABLE IX : Thermal fission of ^{239}Pu, E_β Different irradiation times: 10, 10^2, 10^3, 10^4, 10^5 seconds.

T_{cool} (S)	DEVILLERS	PETROV (Exp.)
1 0	.174 0	.369 0
2	.118	.261
5	.626 -1	.136
1 1	.387	.739 -1
2	.226	.445
5	.104	.190
1 2	.534 -2	.909 -2
2	.247	.439
5	.888 -3	.168
1 3	.453	.810 -3
2	.232	.390
5	.783 -4	.150
1 4	.292	.720 -4
		.250

TABLE X: Thermal fission of ^{239}Pu, Tir = 0, 1 fission, E_γ [MeV s^{-1} $fission^{-1}$]

TCOOL (s)	DEVILLERS	SCOBIE SCOTT (Calc)	SCOBIE SCOTT (exp)
5 0	1.19		1.12
1 1	1.15		1.07
2	1.12		1.01
5	1.08		0.95
1 2	1.05	1.056	0.92
2	1.03	1.035	0.90
5	1.01	1.00	0.89
1 3	1.	.99	0.90
2	.97	.965	0.88
5		.92	.835
1 4	.91	.896	.84
2	.91	.90	.94
5	.97		1.05
9	1.01		.97

TABLE XI : Thermal fission of ^{233}U, $E\beta$, Tir = 10^4 s, P = $\frac{^{235}U}{^{233}U}$

TCOOL (S)	DEVILLERS	BUNNEY-SAM	BUNNEY - SAM / DEVILLERS
1 3	.469 -3	.720 - 3	1.53
2	.242	.370	1.52
5	.830 -4	.102	1.22
1 4	.329	.381 - 4	1.15
2	.116	.180	1.55
5	.329 -5	.541 - 5	1.64
1 5	.137	.210	1.53
2	.552 -6	.809 - 6	1.46

TABLE XII.1 : Fast fission of ^{235}U, Tir = 0, 1 fission
E_γ [MeV s^{-1}]

ΔT_{cool} (S)	DEVILLERS	TOBIAS	FISHER ENGLE (exp)	FISH.ENG / DEVILLERS
0.2 - 0.5	.337 0	.475 0	.564 0	1.67
1 - 2	.199		.311	1.56
4 - 5.5	.933 - 1	.117	.153	1.63
10 - 13	.468	.512 -1	.706 -1	1.50
35 - 45	.151	.177	.221	1.46

^{235}U

ΔT_{cool} (S)	DEVILLERS	TOBIAS	FISHER ENGLE	FISH.ENG. / DEVILLERS
0.2 - 0.5	.244 0	.300 0	.529 0	2.16
1 - 2	.137		.296	2.16
4 - 5.5	.631 -1	.820 -1	.138	2.18
10 - 13	.341	.383	.642 -1	1.83
35 - 45	.126	.146	.197	1.56

^{239}Pu

Table XII.2: Fast fission of ^{235}U and ^{239}Pu, Tir = 0.1 Fission, E_γ [MeV s^{-1} fission $^{-1}$]

	Calculation		Experiment
T_{cool} (days)	DEVILLERS	TOBIAS	JOHNSTON
40		.341 -7	.354 -7
50	.243 -7	.248	.262
60		.192	.205
70	.151	.155	.168
80		.130	.141
90	.110	.112	.121
100		.994 -8	.105
110	.865 -8	.889	.933 -8
120		.734	.753
130	.714		
140		.672	.686
150	.602	.619	.627

TABLE XIII.1 : Fast fission of ^{239}Pu; Tir = 0;
1 fission; $E_\beta + E_\gamma$ [MeV, s^{-1}, $fission^{-1}$]

Tir days	Tcool days	DEVILLERS (WATT)	L O T T (exp) (WATT)	L O T T / DEVILLERS
. 172	. 1606	0.0403	0.0507	1.26
. 456	. 1330	0.204	0.222	1.09
. 589	. 1202	0.267	0.278	1.04
. 1212	. 503	. 799	. 809	1.01
"	. 547	. 730	. 735	1.01
"	. 589	. 675	. 684	1.01
"	. 636	. 621	. 639	1.028
"	. 720	. 542	. 540	1.
. 463	. 326	1.211	1.256	1.04

TABLE XIII.2 : Fast fission of ^{239}Pu (60%) + ^{235}U (40%)

$E_\beta + E_\gamma$

Ratio $\frac{\text{experiment}}{\text{calculation}}$

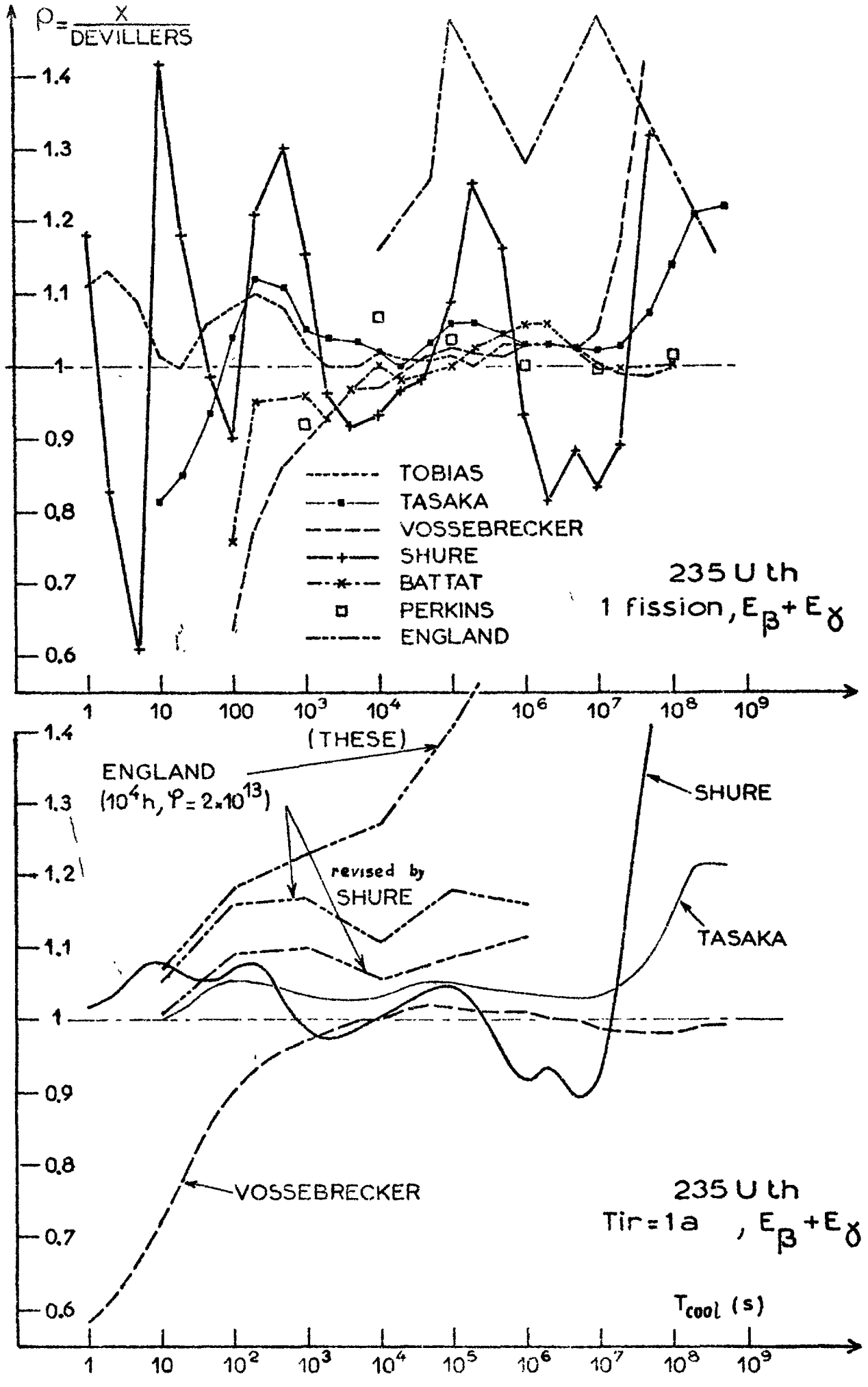

FIG. 1 : ^{235}U th - Comparison of evaluations

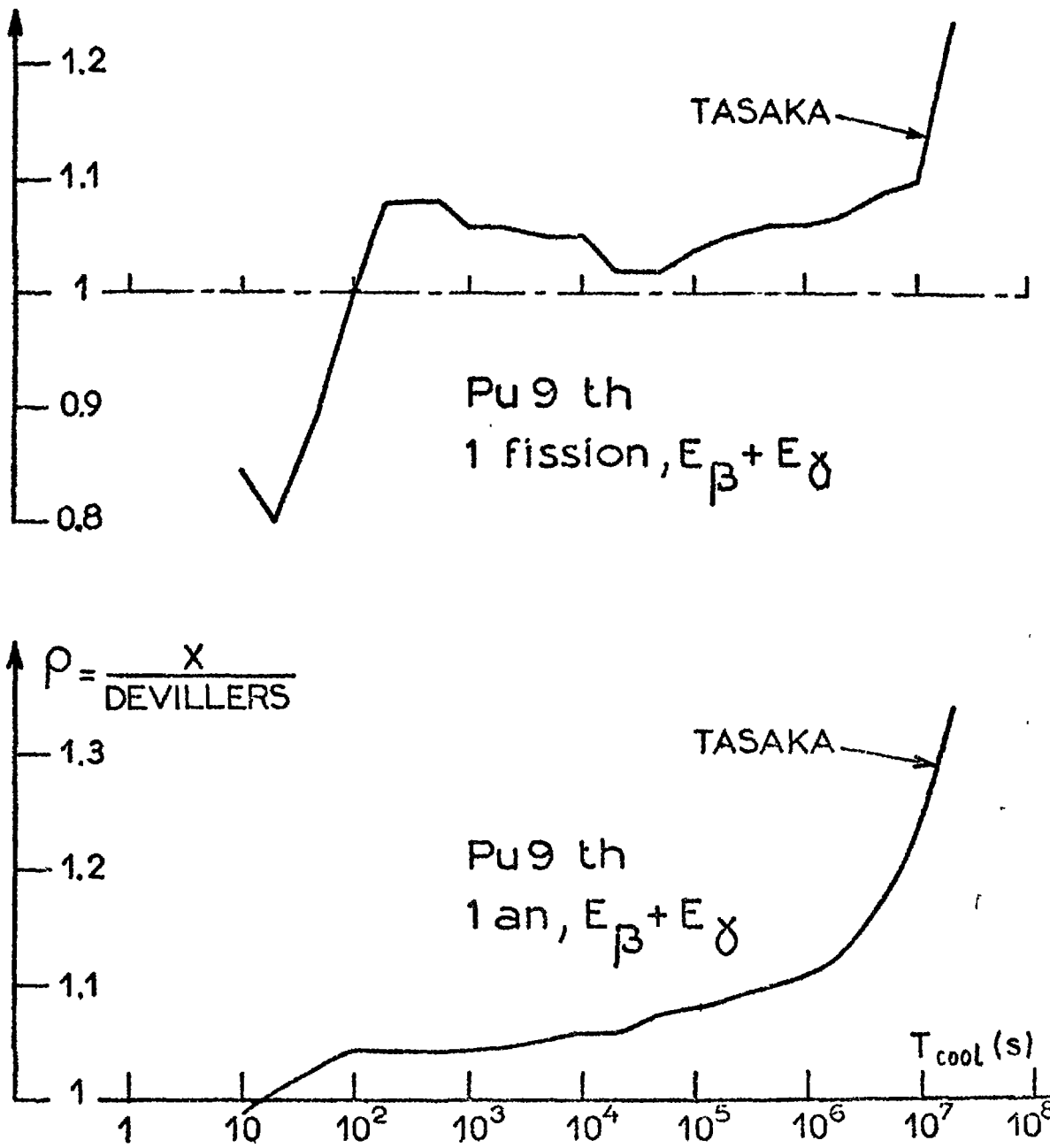

FIG. 2 : ^{239}Pu th - Comparison of evaluations

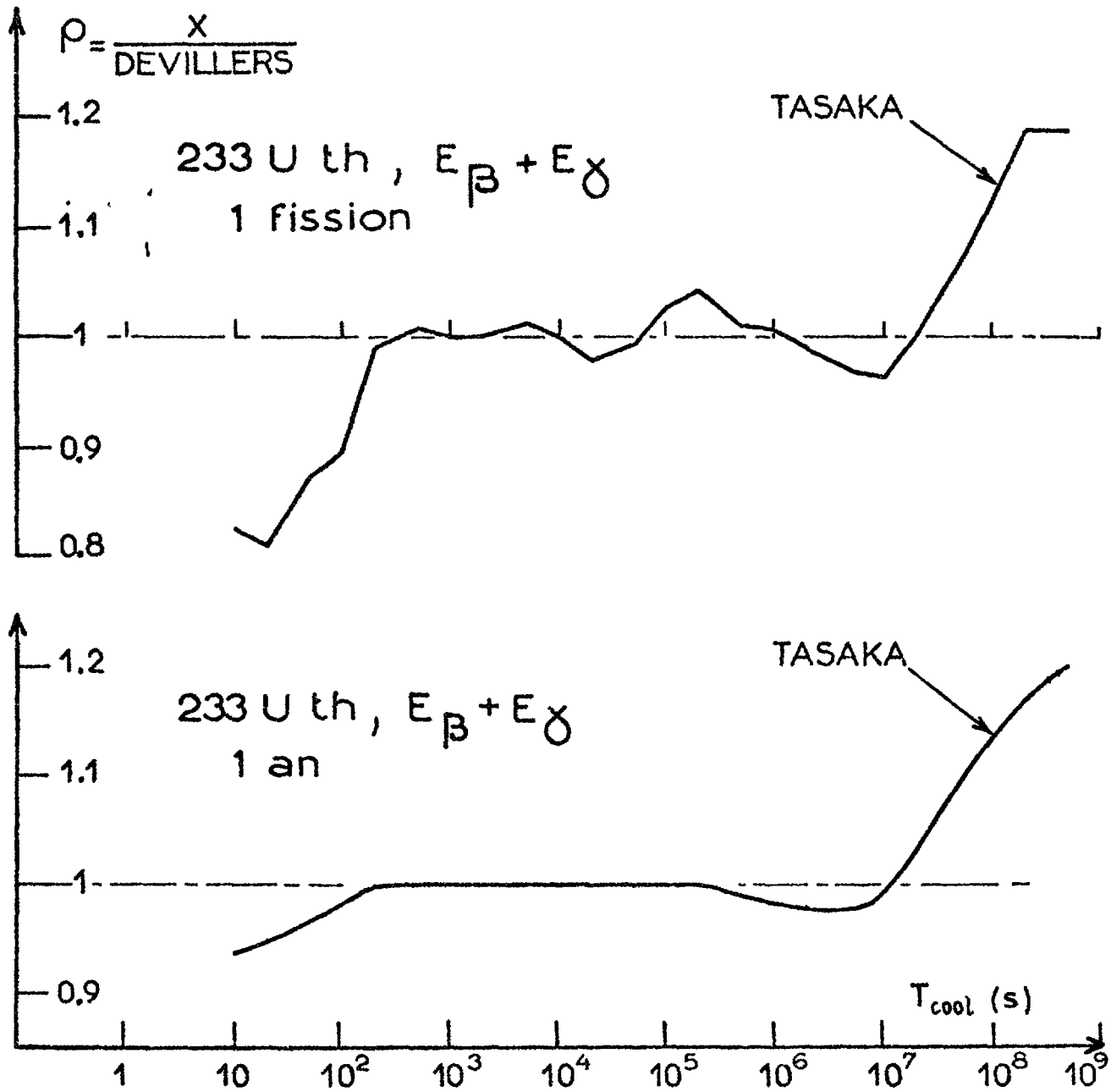

FIG. 3 : ^{233}U th - Comparison of evaluations

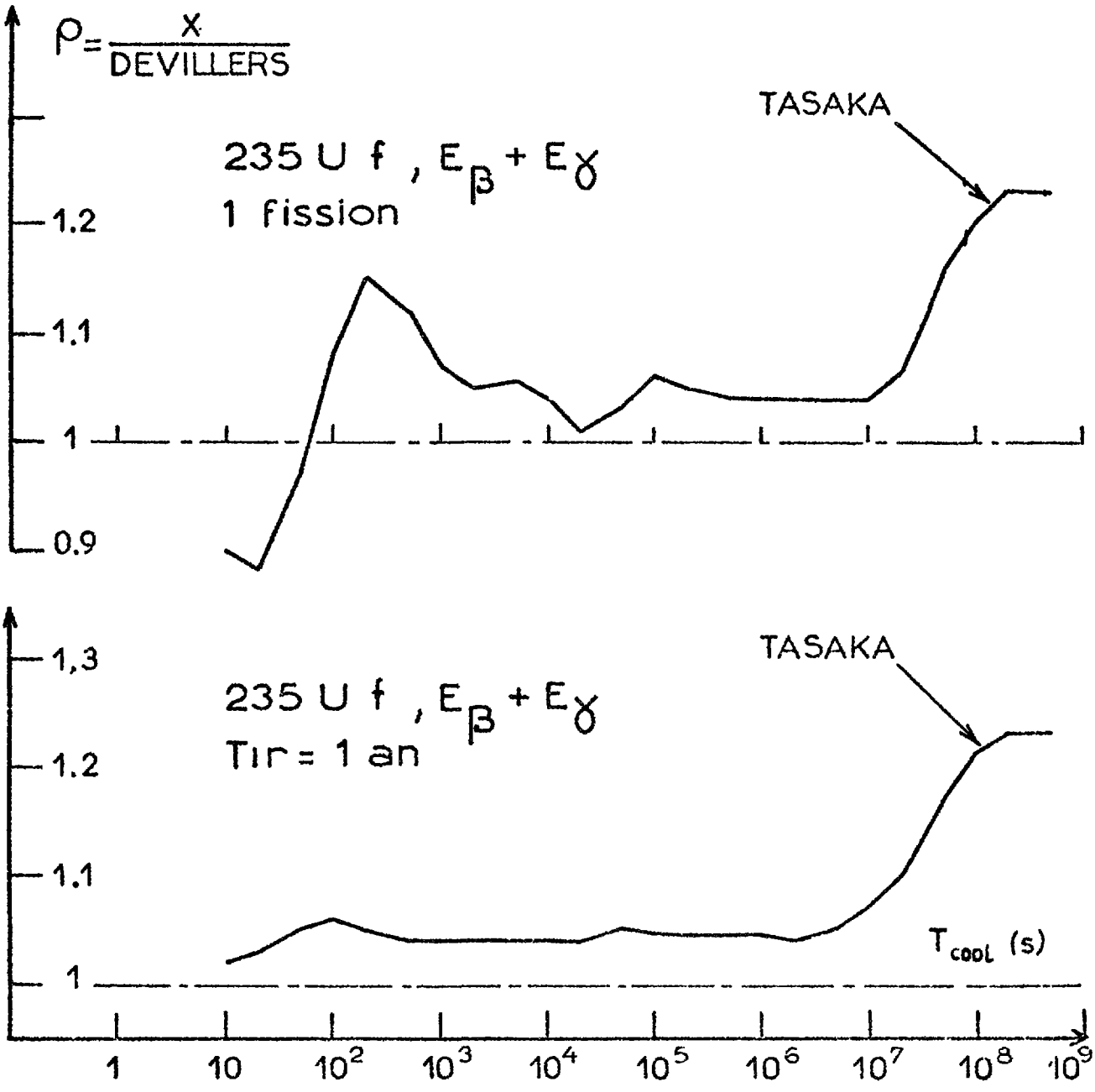

FIG. 4 : ^{235}U Fast comparison of evaluations

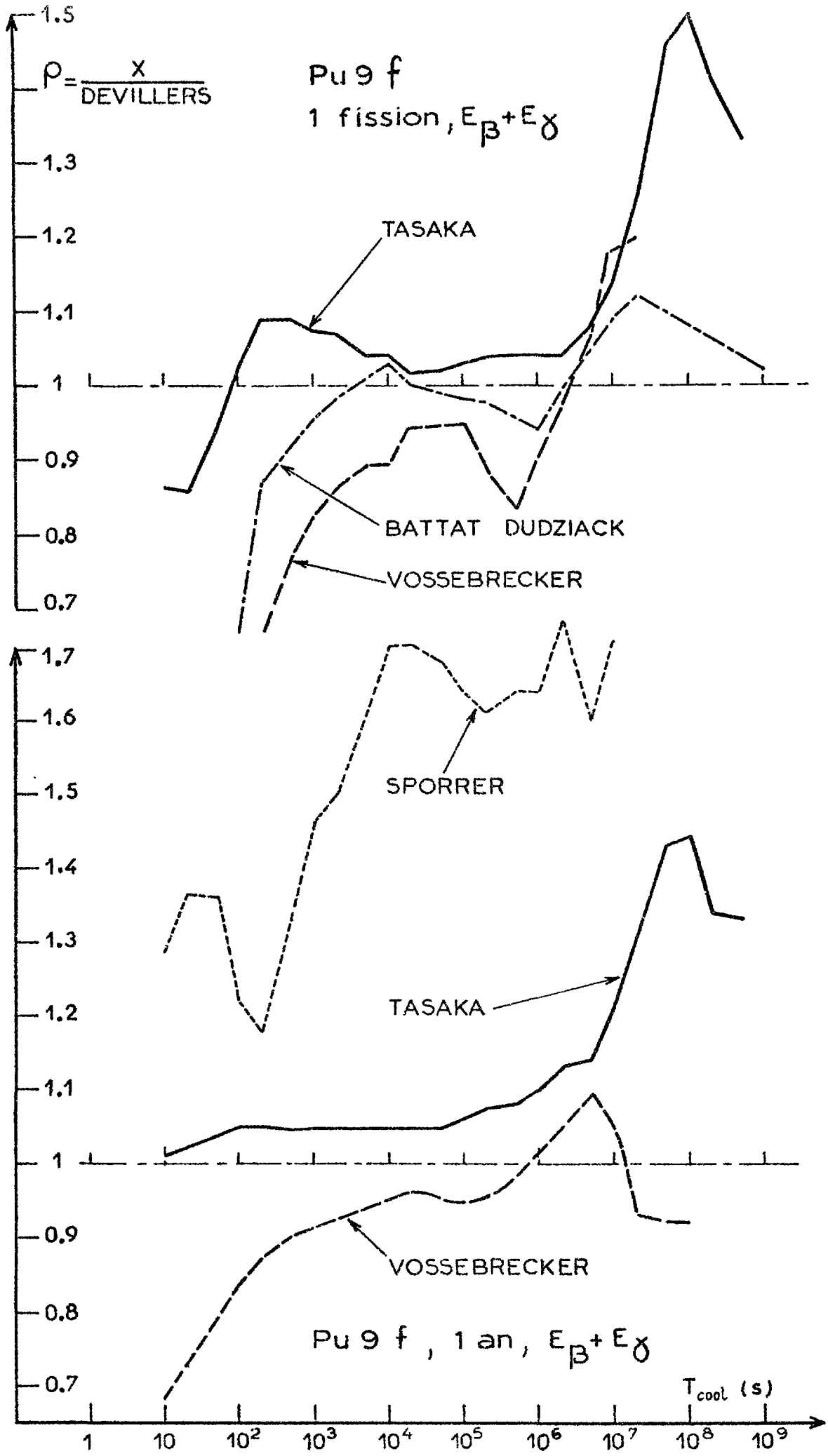

FIG. 5 : ^{239}Pu Fast comparison of evaluations

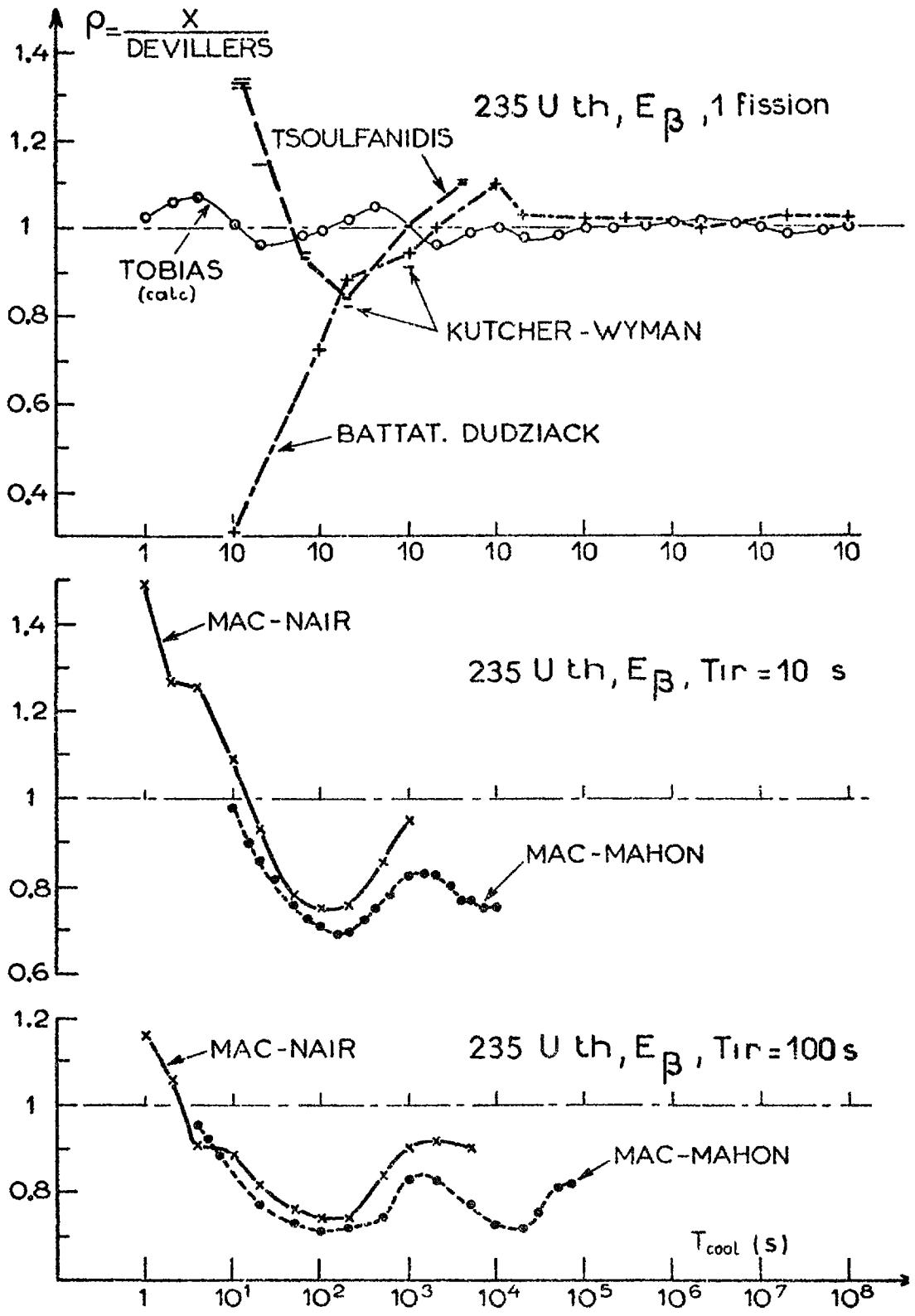

FIG. 6.1 : ^{235}U th, E - Comparison of evaluations and experimental results

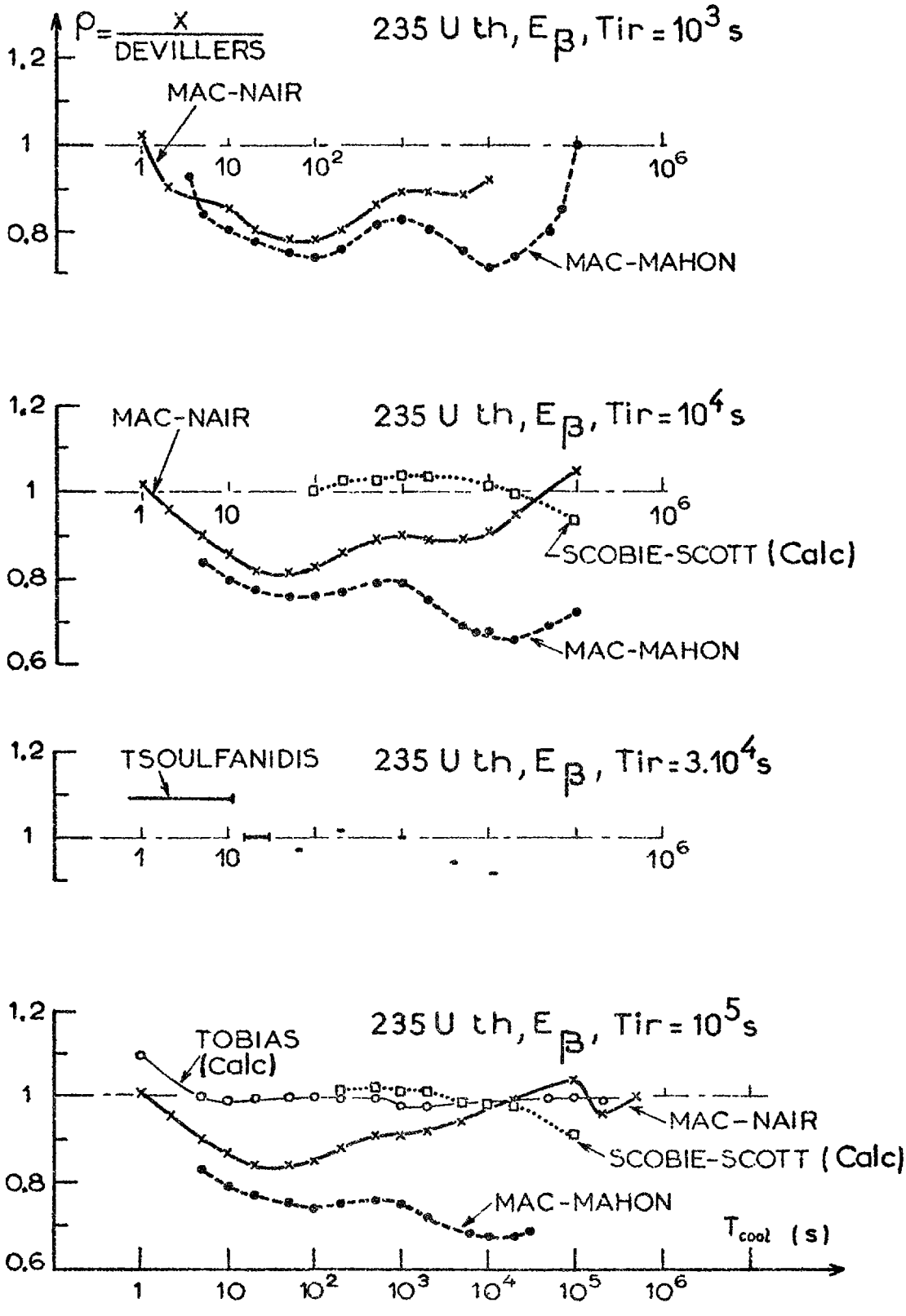

FIG. 6.2 : ^{235}U th, E - Comparison of evaluations and experimental results

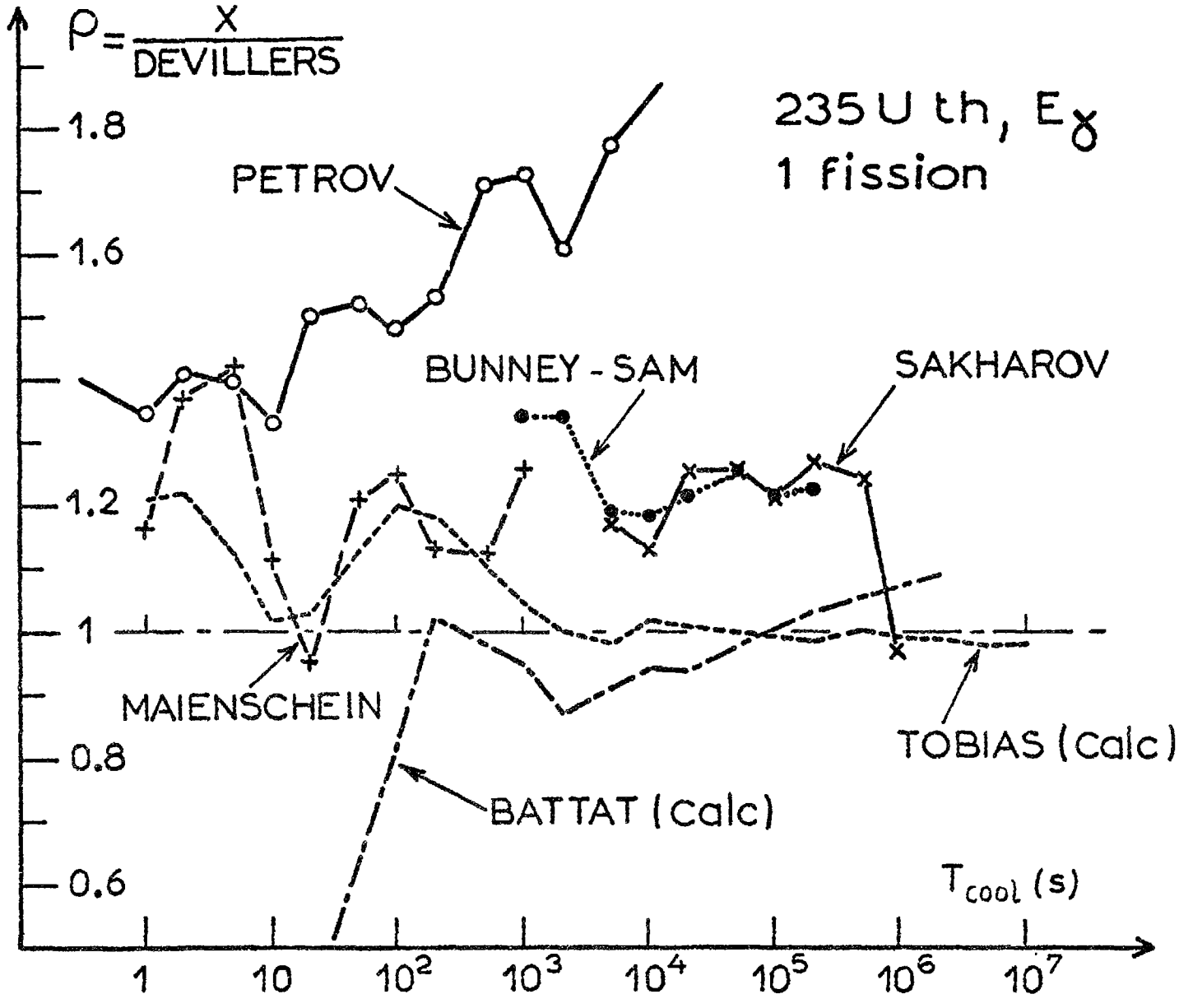

FIG. 7 : ^{235}U th, E - Comparison of evaluations and experimental results

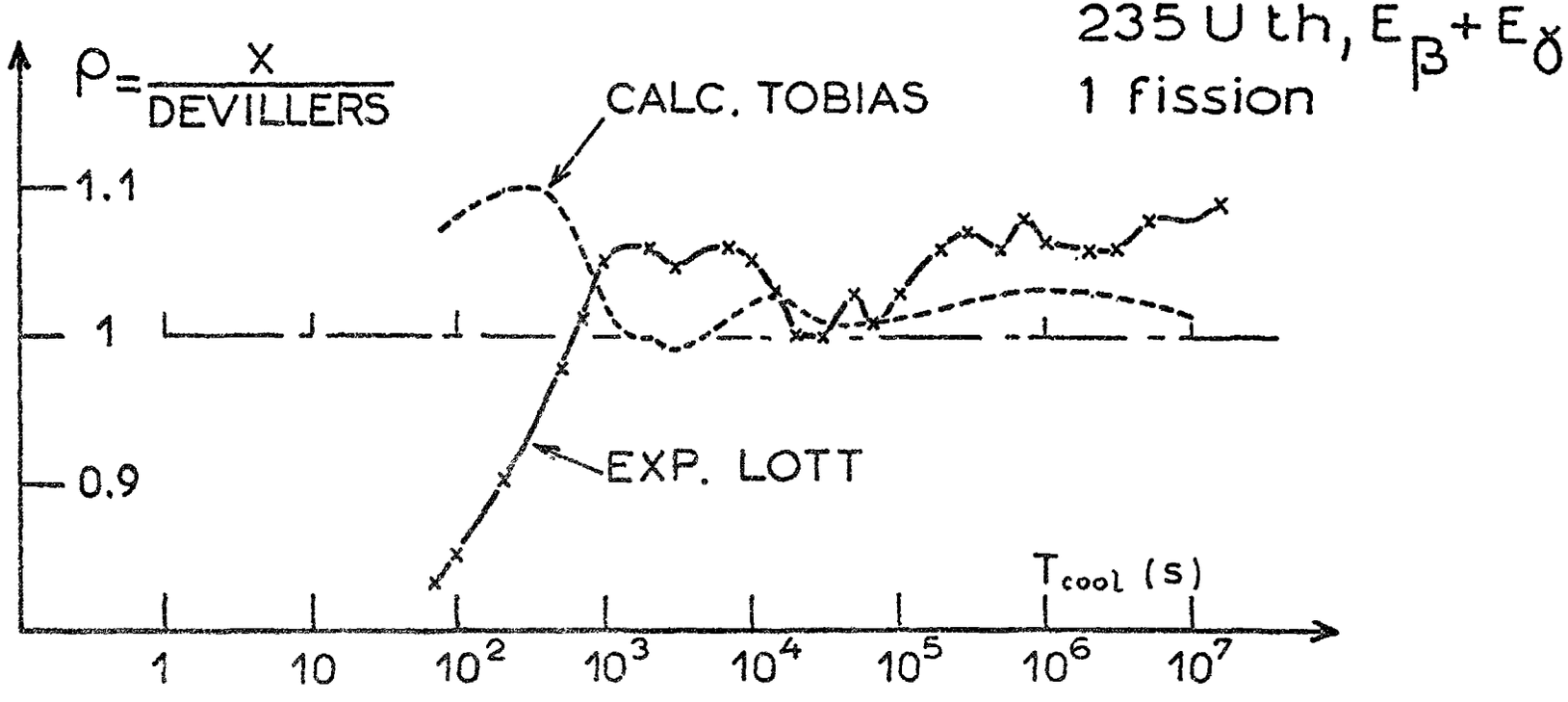

FIG. 8 : ^{235}U th, E - + E Comparison of evaluations and experimental results

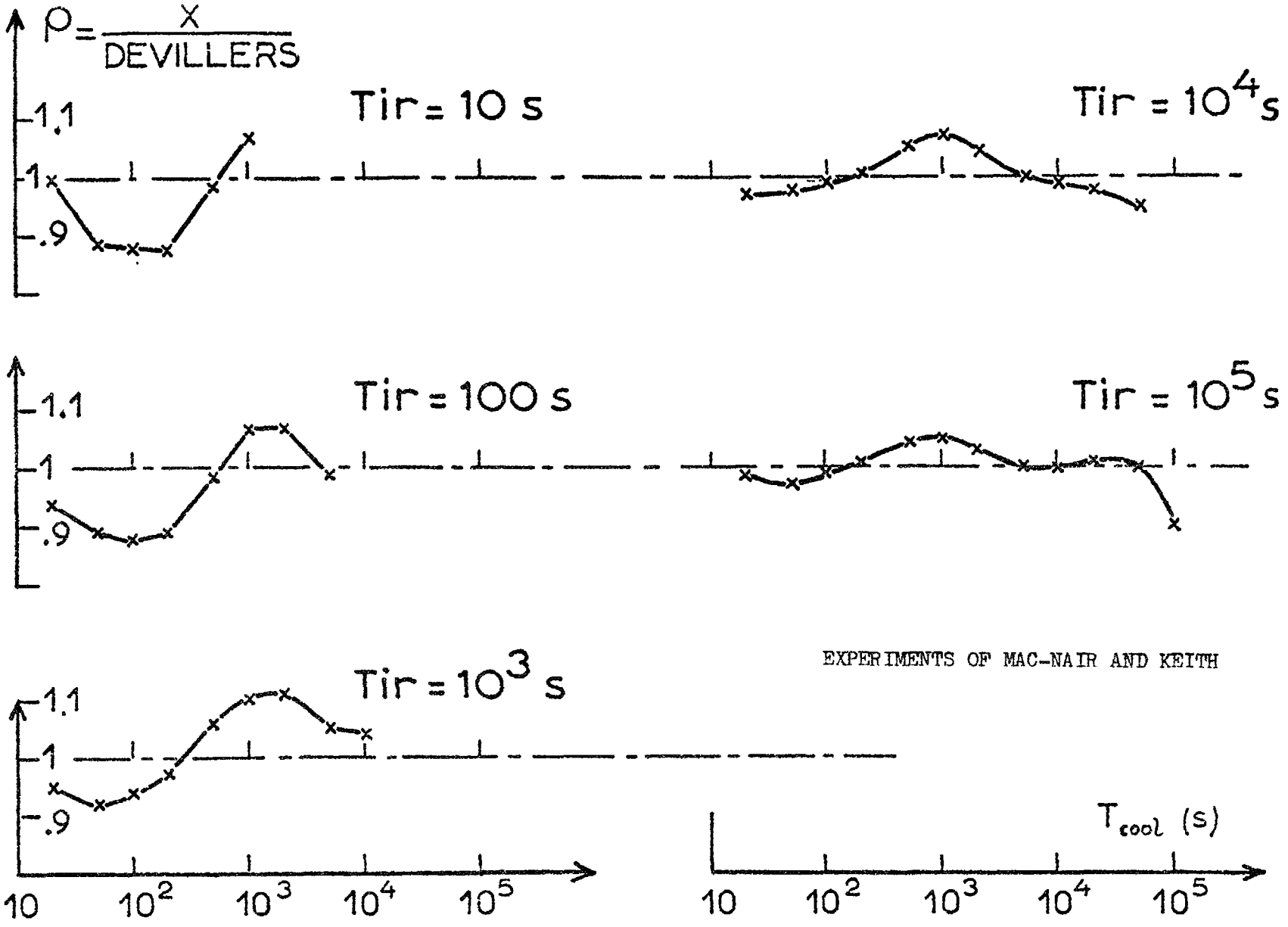

FIG. 9 : ^{239}Pu th, E - Comparison of evaluations and experimental results

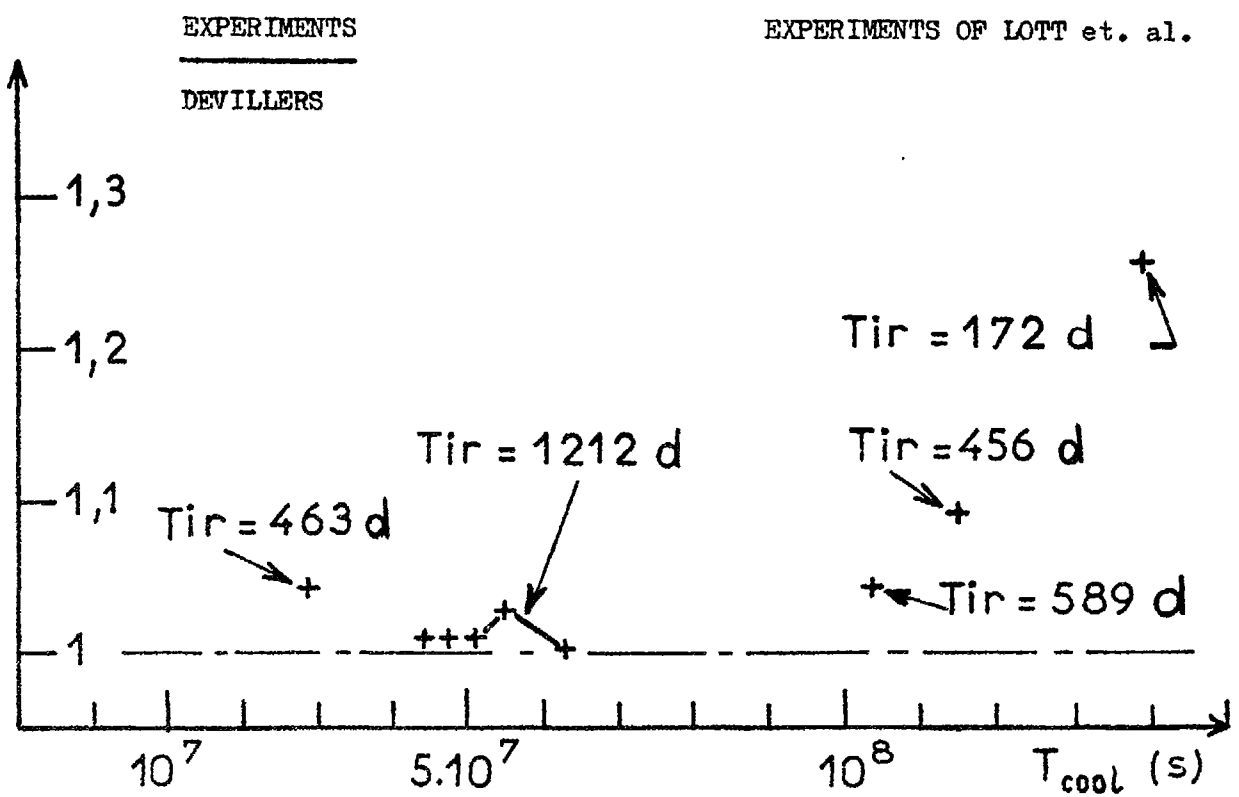

FIG. 10 : ^{235}U fast 40% + ^{239}Pu fast 60% Comparison of evaluation and experimental results

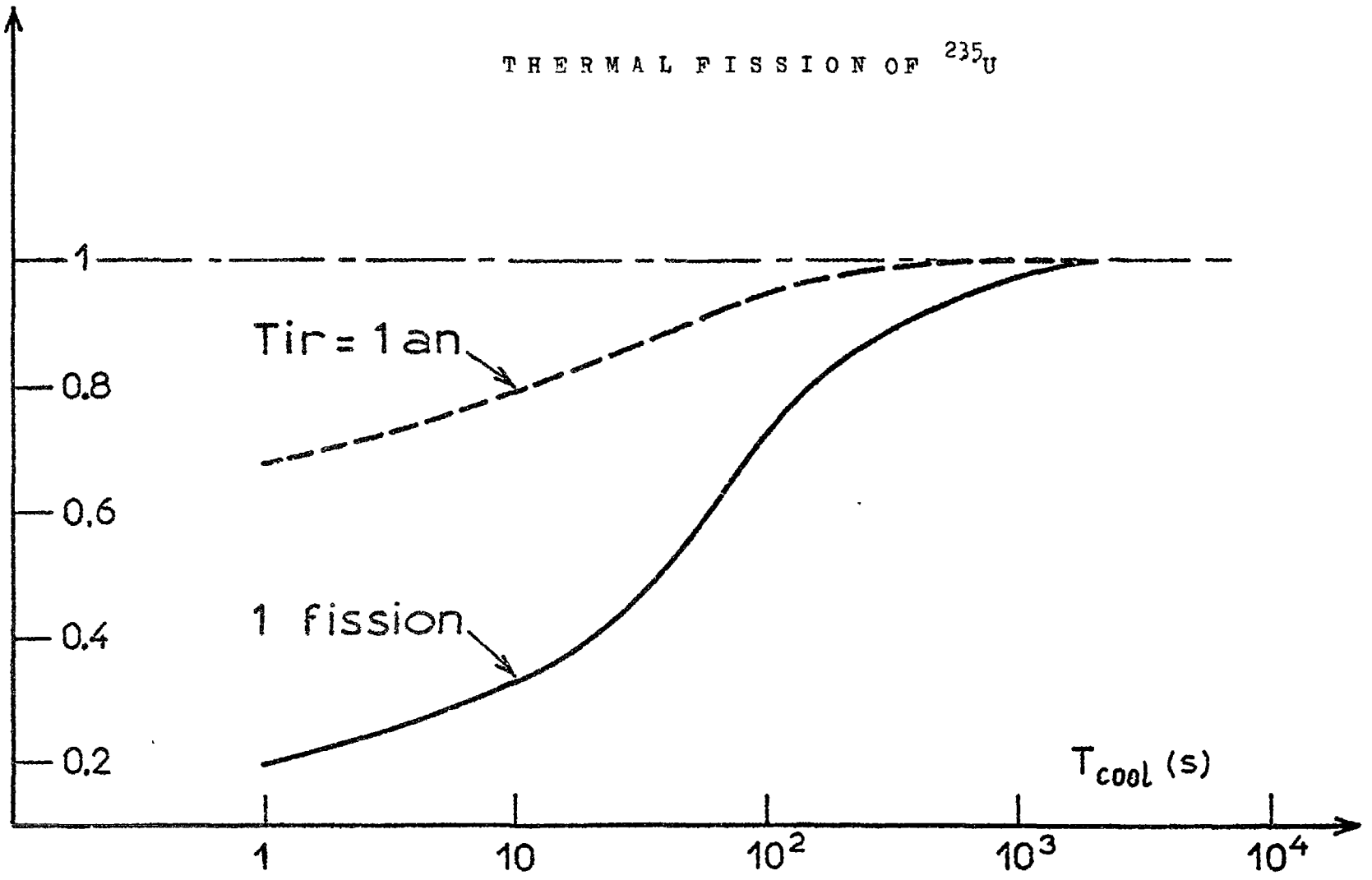

FIG. 11 : Contribution of known fission products to the residual power

Review Paper No. 16:

PREDICTION OF UNMEASURED FISSION PRODUCT YIELDS

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Abstract:

This paper summarises some methods of predicting unknown fission yields. Gussian fitting methods are found to be a possible means of extrapolating fission yields for different neutron bombarding energies and for fissioning species for which measurements have not been made.

Charge division and dispersion are also reviewed and the important enhancement effect for the direct yields of even Z nuclei is demonstrated. However, no universal charge division prescription for all fissioning nuclei is found. The yield enhancement for even Z nuclei is shown to be partly responsible for the fine structure in the mass yield curves.

A new evaluation of prompt neutron remission for ^{235}U is made for use in the calculation of charge division for this nucleus.

1. INTRODUCTION

Strutinksy's [1-3] insight into the role of shell structure in deformed nuclei has been used by a number of authors to gain new understanding of fission [4-6]. Others [7,8] have used a semi-empirical shell correction of Myers and Swiatecki [9] with encouraging agreement with the qualitative features of asymmetric mass division. The quantitative predictions of current theories, however, would not be good enough for use in design and burnup calculations (for example) and where more reliable data are required resort must be made to a considerable folklore of fission systematics. Briefly then, how systematic is fission?

Perhaps the most striking feature of the mass distribution in low energy fission is the great stability over a wide range of fissioning nuclei of the heavy mass peak [10] and of the steeply rising section of the yield curve near the mass number 132. The mean heavy fragment mass both before and after the emission of prompt neutrons is practically constant over a range of some 30 in

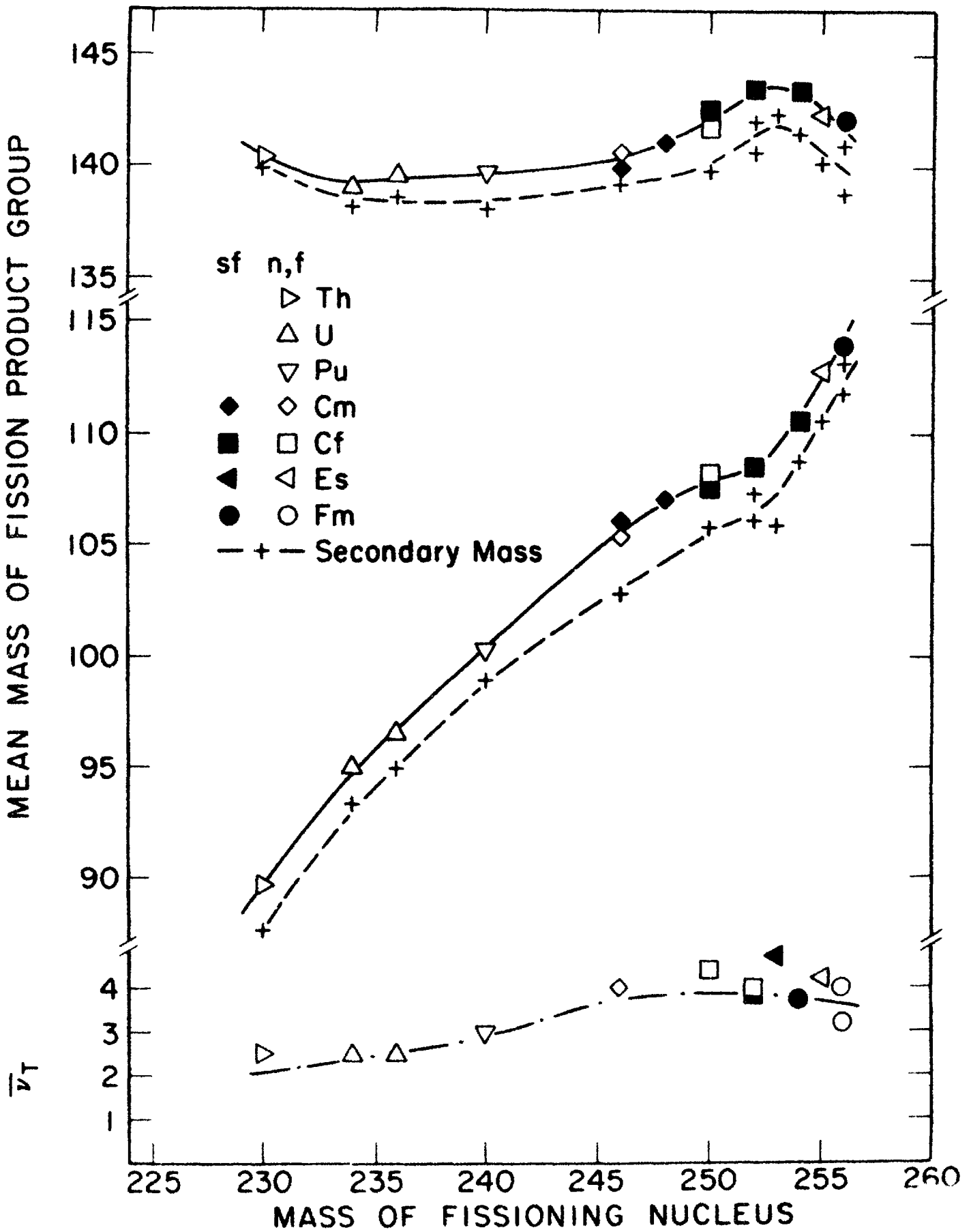


FIGURE 1 Average primary masses (solid curves) and post neutron emission masses (broken curves) of the fission product groups. Also shown is the average $\bar{\nu}_T$ as a function of the mass number of the fissioning nucleus. (From [12]).

fissioning nucleus mass numbers [11]. Recent work [12], however, indicates the presence of some structure between mass numbers 250 to 255 as shown in Figure 1. Also plotted in Figure 1 are values of $\bar{\nu}_T$, the average number of prompt neutrons emitted in fission.

The widths of the mass peaks also vary in a systematic way with fissioning mass number. A plot of the full width at one tenth maximum [11] shows in fact a linear increase with increasing mass. A more complicated function of mass number and excitation energy above the fission barrier is the peak to valley yield ratio. As shown in Figure 2 for thermal neutron induced fission, the symmetric valley fills up quite rapidly with increasing mass number until, accerding to John et al. [13], predominantly symmetric fission occurs for thermal neutron fission of ^{257}Fm. For each fissioning system, symmetric fission becomes increasingly favoured as excitation energy increases [14,28] and even for the energies involved in reactor-neutron induced fission we can see a significant symmetric enhancement.

Figures 3 and 4 [12] give the measured primary (pre-neutron emission) mass distributions for a number of spontaneously fissioning systems and others fissioning by thermal neutrons. The shaded vertical bars indicate the fragment masses at which even Z charges are calculated to occur. The considerable fine structure occurring in the primary mass distributions correlates very closely with the even Z fragments and heavy mass groups containing 52 to 58 protons are particularly enhanced, the more so when in conjunction with a light fragment of 42 protons. Mass distribution fine structure has previously been observed in mass versus kinetic energy studies [15-17] and is particularly evident at low excitation energies where again it appears to be a result of preferential formation of even-even primary fission fragments. The fine structure carries through to the post neutron emission mass distribution where it appears as an enhancement of fission products with even Z [18]; most of the even neutron number fine structure having been washed out in the neutron emissions.

2. DEFINITIONS

The following definitions follow closely those used for the EXFOR library as outlined in the LEXFOR manual (see also CINDA, Vol. 1, page XIII and Vol. 2 page A5 (Annex)).

A primary fission fragment (initial or pre-neutron emission fragment) is formed at the time of scission. Usually these are the two massive products of binary scission, but one can also have in accompaniment a scission neutron (in perhaps 25 per cent of fissions) or a light charged particle (<1 per cent of fissions). Rarely, break up into three comparable mass fragments occurs. After rapidly ($< 4 \times 10^{-14}$ sec) losing prompt neutrons and γ-rays ($< 10^{-11}$ sec) the fragments are called secondary or post neutron emission fragments or primary fission products. In practice, scission neutrons are not distinguished from the later prompt neutrons and all will be assumed to arise from the fully accelerated fragments.

The primary products then slowly ($>10^{-2}$ sec) decay via β decay forming the secondary products of a decay chain until eventually the stable end product is reached.

The independent (direct) fission yield is the probability (atoms/fission) that a particular nuclide (Z,A) is formed in fission following prompt neutron emission, but before any β decays. Usually this is quoted as a fraction of the cumulative chain yield of that isobar.

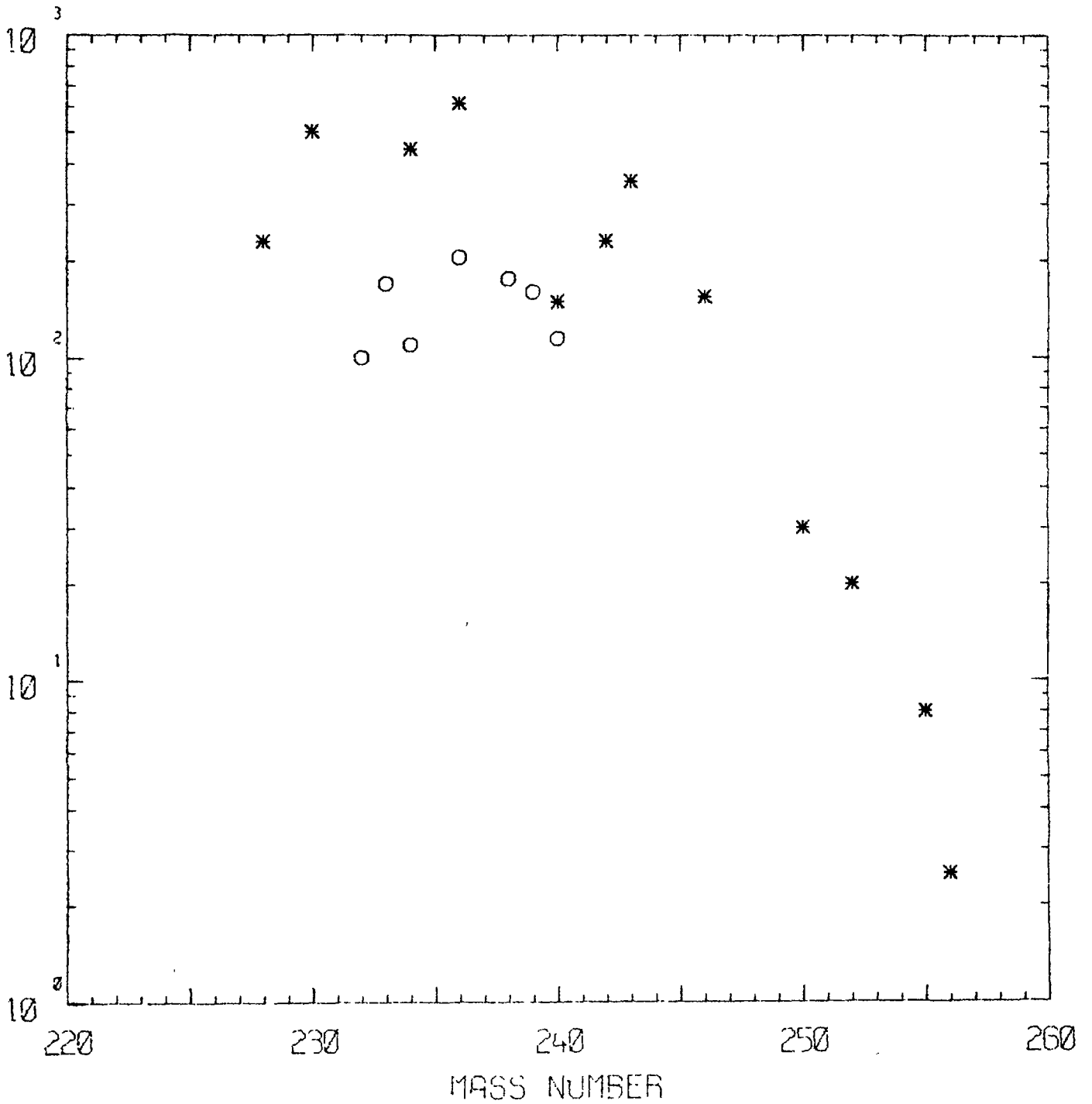


FIGURE 2 Peak/Valley mass yield ratio versus mass number of the fissioning nucleus.

* - thermal neutron fission
O - reactor neutron fission

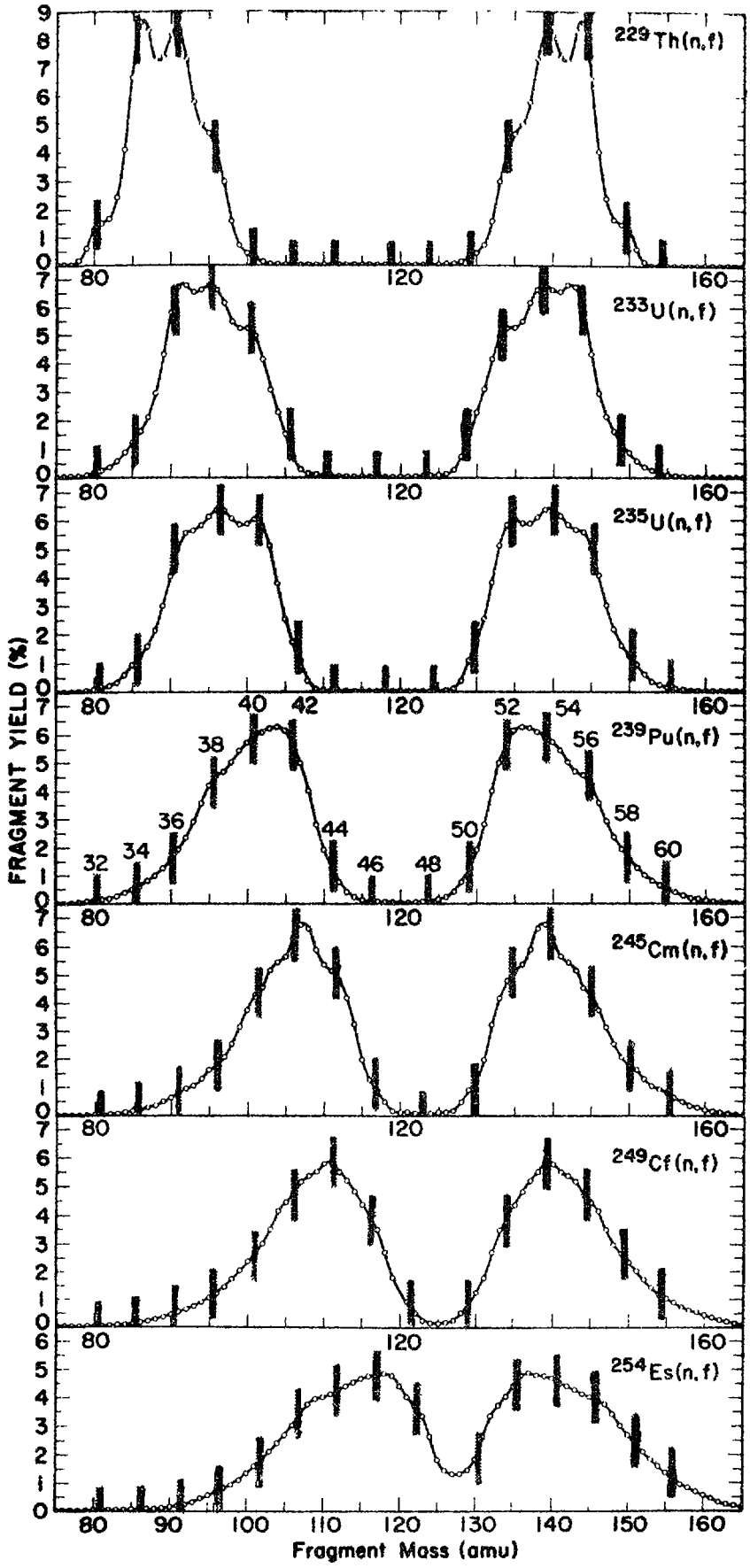


FIGURE 3 Primary fragment mass distributions obtained for thermal-neutron induced fission [12] corrected for mass resolution. The shaded vertical bars indicate calculated positions of even Z fragment masses.

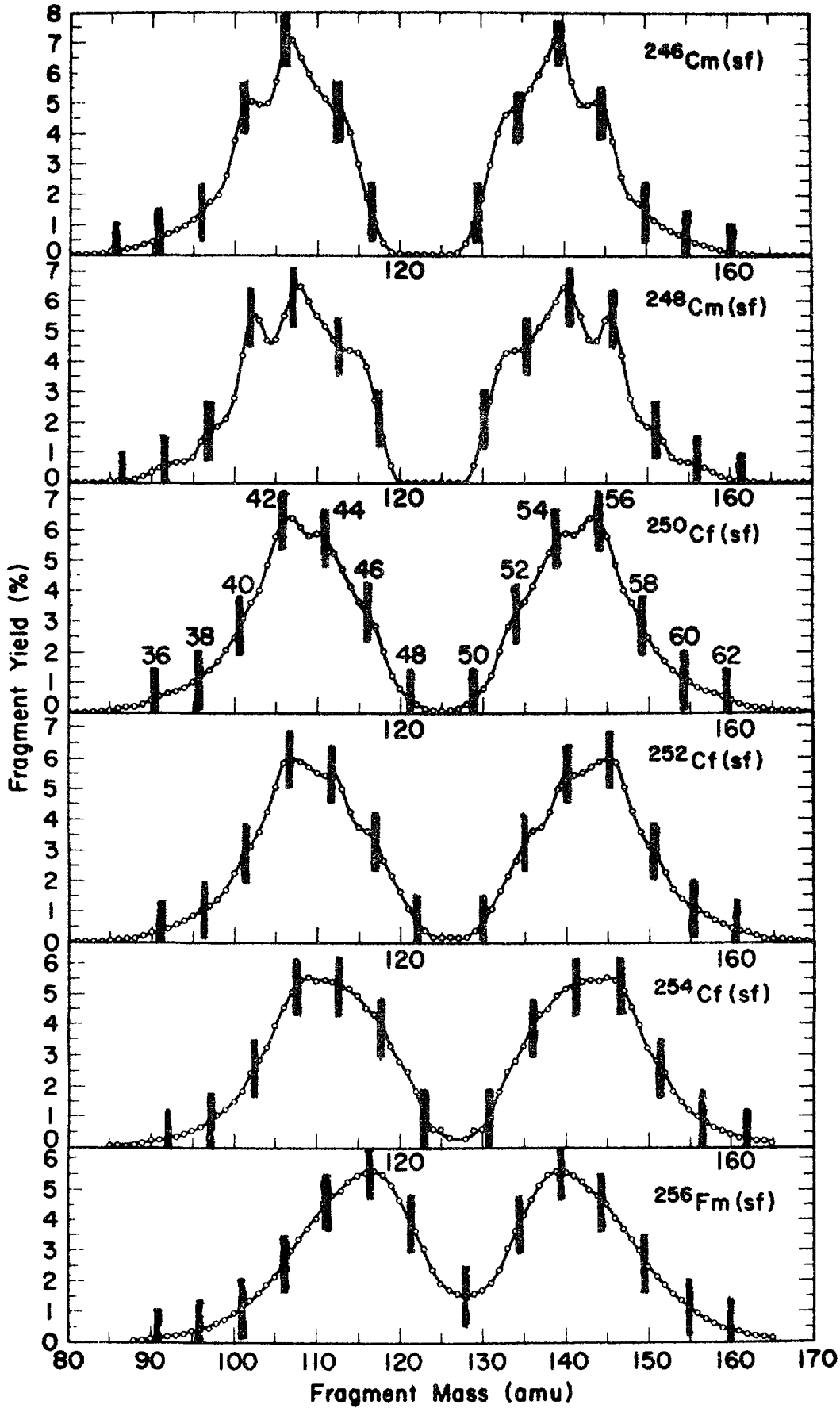


FIGURE 4 Primary fragment mass distributions obtained for spontaneous fission [12].

The cumulative yield of a fission product is the sum of the independent yield of the nuclide in question and those of all its β-chain precursors plus rarely a contribution from delayed neutron emission. This is usually quoted as a fraction of the cumulative chain yield of that isobar.

The cumulative chain yield is just the cumulative yield of the β-stable nuclide at the end of the decay chain, while the total chain yield also includes any additional independent yields of shielded nuclei (with Z > Z stable). The sum of the total chain yields is 2.00.

Since the mass number does not change during β-decay, the final mass distribution is determined after the emission of prompt neutrons (except for some slight corrections due to delayed neutron emission).

3. FILLING IN PARTIALLY MEASURED MASS YIELD CURVES

The experimentally measured final mass yield curves still contain gaps for which yields must be estimated. Even for ^{235}U thermal neutron induced fission, the most extensively studied case, 0.6 per cent of the light masses and 0.1 per cent of the heavy masses must be interpolated [19]. For the thermal neutron fission of ^{233}U the figures are 1.3 per cent and 5.6 per cent; for ^{239}Pu 6.9 per cent and 5.3 per cent, while in the case of ^{241}Pu nearly 10 per cent of the heavy yield curve and 40 per cent of the light mass curve relies on interpolation. When one looks at fission yields from reactor neutron induced fission, the situation is generally worse and it is therefore desirable to provide a sound method of predicting unknown yields. At present, the favoured method used by most evaluators is interpolation by means of a smooth curve drawn through experimental points [19-21]. This method is adequate where the experimental gaps are not too wide, but is open to question where fine structure is expected or where large gaps in the experimental data exist. The final yields satisfy the condition that

$$\sum_A Y(A) = 2.0 \quad ,$$

where Y(A) is the final yield for mass number A.

However, this sum is usually taken separately over both light and heavy mass groups [19] with the division between the two sums at $\bar{A}$, the mean nucleon number given by

$$\bar{A} = (A_F - \bar{\nu}_T)/2 \quad ,$$

where A_F is the mass number of the fissioning nucleus and $\bar{\nu}_T$ the average number of prompt neutrons emitted per fission. Of course this condition is of little use in obtaining unknown yields and in practice is used to normalise the data after the missing yields have been interpolated. A more powerful boundary condition restricts the shapes of the final mass peaks such that

$$\sum_A Y(A) \cdot A = 2\bar{A} \quad ,$$

where the sum is taken over both light and heavy mass groups [19]. The application of this boundary condition serves to adjust interpolated yields, but may in principle allow a decision to be made between very discrepant measured yields [22]. Note that mere satisfaction of the boundary conditions does not guarantee that the yields are correct, however clearly the converse is applicable and occasionally evaluated mass yield curves in the past have not satisfied the second condition above [22,62]. Note, however, that some experimental error attaches to both sides of this equation.

When large portions of the yield curve are unmeasured, as for example the ^{240}Pu fission yield in fast reactors where no data are available, one must attempt an interpolation between, or extrapolation from, the measured mass yield curves.

Sidebotham [23] does this in the most straightforward way. He assumes that the mean widths of the light and heavy mass peaks do not vary with mass number and furthermore, that the peaks of individual isotopes in known and unknown curves are merely displaced along the mass axis. Then by bodily shifting a known mass yield curve and using the fact that the primary (pre neutron emission) mass distribution can be mirrored about the symmetric mass point, the unknown final mass yield can be constructed. In estimating the 'mirror' yield the number of neutrons emitted by each fragment is calculated from the simple formulae of Terrell [24]. For the derived yields Sidebotham [23] estimates about 20 per cent error in the peaks and maybe 10 per cent in the flanks of the unknown yield distribution. Of course in some cases, several known mass yields may be used to form the basis of the extrapolation and in such cases Sidebotham [23] reports a good degree of agreement between the various estimates. Some examples of Sidebotham's [23] calculations are given in [63]. It can be seen that the agreement with experiment is in general rather good.

It is not our intention to discuss in any further detail the methods outlined above. Instead, let us ask what it might be possible to achieve in predicting unknown yields from a knowledge of the systematics of fission. We defer until a later section any discussion of fine structure in the yield curves, which as we indicated earlier, appears to be a result of enhanced formation probability for even-even fragments.

Let us assume that a suitable parameterisation of the primary structureless mass yield curve (i.e. the mass yield curve with fine structure smoothed out) can be found. Then one might reasonably expect to find a smooth variation of the parameters describing the fit as the mass of the fissioning nucleus altered and as a function of incident neutron energy. Then, following the method suggested by Crouch [25], the final mass distribution can be generated:

$$Y(M) = y(M)\,P_0(M) + y(M+1)\,P_1(M+1) + y(M+2)\,P_2(M+2) + \ldots ,$$

where $y(M)$ is the primary yield of mass M, and

$Y(M)$ the yield after neutron emission has taken place.

$P_\mu(M)$ is the probability that the fragment of mass M emits exactly μ neutrons:

$$\sum_{\mu=0}^{\infty} P_\mu(M) = 1 .$$

Since little is known about $P_\mu(M)$ for particular values of M, an assumption must be introduced, and Crouch [25] assumes the distribution to be a Poisson distribution:

$$P_\mu(M) = \frac{\exp(-\bar{\nu})\cdot(\bar{\nu})^{\mu}}{\mu!} ,$$

where $\bar{\nu}$ is the mean number of neutrons emitted by the fragment mass M.

In Crouch's treatment $\bar{\nu}$ is assumed known from measurement or alternatively from a semi empirical formula [24].

One then attempts to fit the experimental chain yields Y(M) with an assumed functional form y(M) for the primary mass yield curve. Originally, Crouch [25] suggested that each peak in the mass yield curve might be represented by a simple Gaussian. A further contribution is then required to fit the yields in the symmetric valley and another Gaussian is introduced to take care of this problem. Phenomenologically, this central Gaussian grows larger with increasing energy of the bombarding neutron and has led several authors to describe the process in terms of the so called 'two mode' hypothesis [26].

Unfortunately, a single Gaussian gives a poor fit to the primary mass yield curve and in later work Crouch [27] reports that he is using two Gaussians to describe each peak of the primary mass curve. The means and variances of the Gaussians appear to vary in a systematic way with the mass number of the fissioning nucleus. For example, the means of the two Gaussians fitting the heavy mass peak remain constant while those fitting the light mass peak increase linearly with mass number. Crouch [27] reports that the fit obtained is quite good being within ± 0.2 per cent (standard deviation) on average. Thus the high yields of an unknown mass distribution can be predicted with some degree of confidence from these systematics. Crouch [27] reports that work is proceeding along these lines, but unfortunately, no further details were available for this review.

We have proceeded in a much more simple minded fashion by attempting a fit to the post neutron emission yields directly. This has the advantage of not requiring a postulate about $P_\mu(M)$ nor any advance knowledge of $\bar{\nu}$ as a function of mass number. This advantage is offset by the fact that while the primary mass distribution is strictly symmetric about $A_F/2$, the final yields are not symmetric about $(A_F-\bar{\nu})/2$. However, Walker [19] has shown that at least the high yield sections of the curve are approximately reflection symmetric. At first, we attempted to fit the final (structure removed) yields with three Gaussians (one for each peak plus a symmetric contribution). As expected, the fits were poor at thermal energies, but the quality of the fits improved dramatically as neutron bombarding energy increased. Figure 5 gives the fit to ^{235}U bombarded by 14 MeV neutrons. The mean error is 11 per cent which is typical of other fits at this energy. The parameters describing the fits appeared to be systematic functions of mass number and energy, for example, the Gaussian variance increased linearly with mass number and also with bombarding energy (although in this case only 3 points were available).

In an approach somewhat similar to that used by Crouch, we also tried fitting the final mass distribution to a calculated primary mass distribution minus Terrell's [24] neutron numbers. This separate fit is shown as a dashed line in the figures. However, by using this method, we found no gain in the quality of fit.

Next, each peak in the mass yield curves was fitted with a double Gaussian (assumed reflection symmetric about $\bar{A}$, the mean mass) plus one central Gaussian:

$$Y(A) = N_1\sqrt{\frac{\alpha_1}{\pi}}\left[\exp\left(-\alpha_1(A-\bar{A}-D_1)^2\right) + \exp\left(-\alpha_1(A-\bar{A}+D_1)^2\right)\right]$$

$$+ N_2\sqrt{\frac{\alpha_2}{\pi}}\left[\exp\left(-\alpha_2(A-\bar{A}-D_2)^2\right) + \exp\left(-\alpha_2(A-\bar{A}+D_2)^2\right)\right]$$

$$+ N_3\sqrt{\frac{\alpha_3}{\pi}}\exp\left(-\alpha_3(A-\bar{A})^2\right)$$

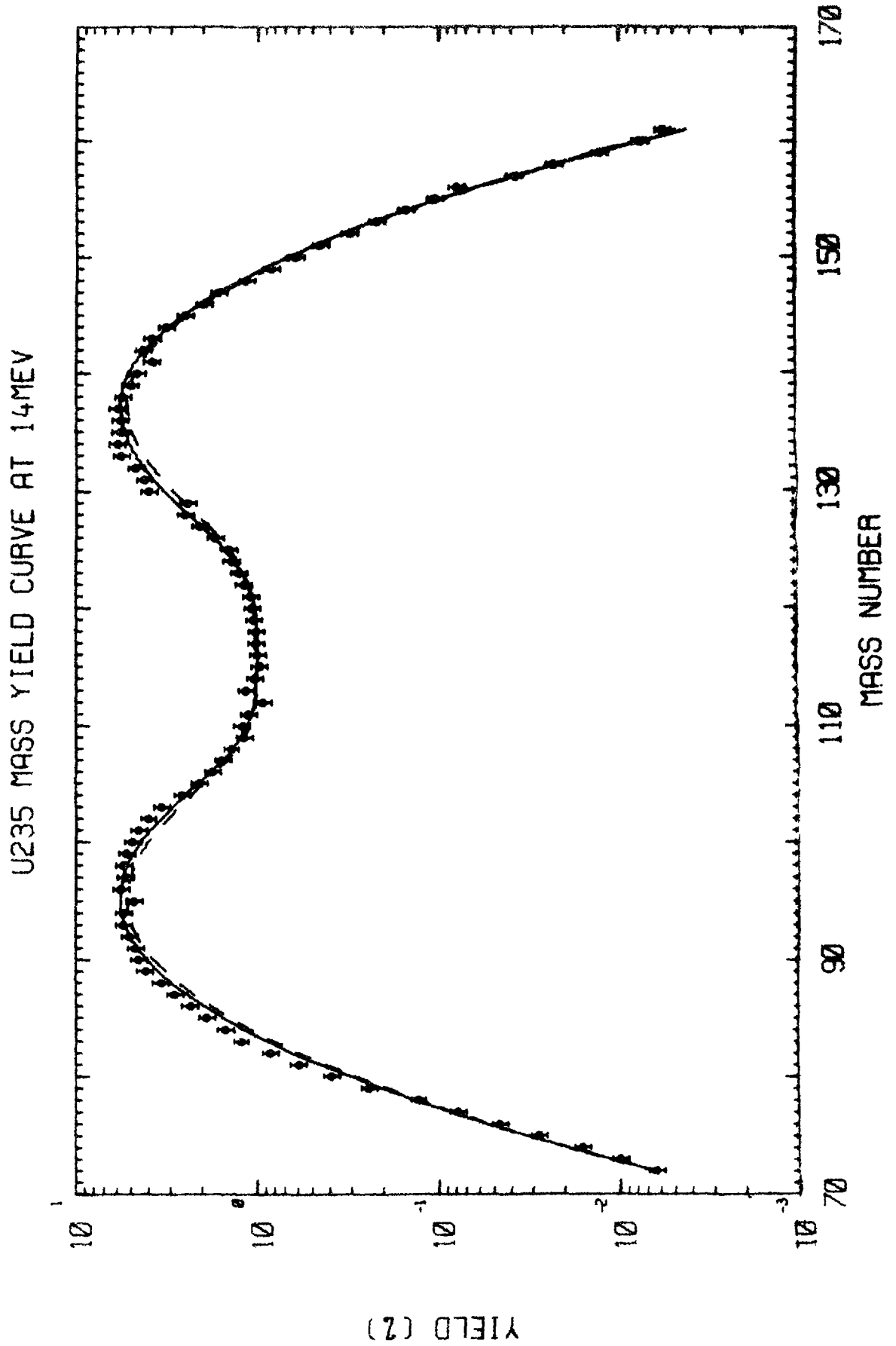


FIGURE 5 Calculated fit to the mass yield curve of ^{235}U + 14 MeV neutrons using 3 Gaussians.

The variances of the Gaussians are given by:

$$\sigma_i = \frac{1}{\sqrt{2a_i}}$$

and the yields were normalised to 200 per cent with

$$N_3 = 2(1-N_1-N_2)$$

Figures 6 and 7 show the fit produced to ^{233}U, ^{235}U, ^{239}Pu and ^{241}Pu fissioning with thermal neutrons. The parameters describing the fits are given in Table 1 for thermal neutrons and fission neutrons. To extrapolate to unmeasured mass yields we fitted the best straight lines to the fitted parameters as a function of mass number. Figure 8 shows a comparison of our calculated ^{240}Pu mass distribution for a fast reactor neutron spectrum compared with Sidebotham's [23] calculation. The overall agreement is rather good and it will be of great interest to learn if Crouch has attempted this calculation yet.

This method can also be used to fill in gaps in a measured mass distribution by fitting to the experimental points if there are sufficient for the fitting procedure to converge. Although our fit to the ^{241}Pu mass distribution was by far the worst of the thermal neutron cases, we give in Table 2 a comparison of the calculated yield at a few mass numbers compared with values interpolated linearly by Flynn and Glendenin [21] and by Walker [19]. The inclusion of fine structure could improve the calculated values.

TABLE I

FITTED PARAMETERS FOR FIVE GAUSSIAN FIT

Nuclide	σ_1	σ_2	σ_3	$\bar{A}$	D_1	D_2	N_1	N_2
			(a) Thermal Neutrons					
^{233}U	4.475	2.326	11.78	116.27	23.7	16.2	0.829	0.169
^{235}U	4.260	3.214	10.10	118.34	26.2	17.5	0.574	0.424
^{239}Pu	5.481	2.934	4.09	118.64	22.5	15.5	0.595	0.403
^{241}Pu	3.664	4.562	3.74	121.34	30.7	18.3	0.135	0.864
			(b) Fission Spectrum Neutrons					
^{232}Th	3.320	2.795	11.41	115.72	26.9	20.1	0.636	0.356
^{233}U	4.420	1.470	13.87	116.03	23.6	16.8	0.878	0.109
^{235}U	4.652	2.784	12.41	117.03	23.6	16.3	0.766	0.228
^{238}U	4.910	2.026	9.82	118.54	22.7	15.4	0.769	0.228
^{239}Pu	5.482	3.702	12.63	118.40	23.1	16.6	0.434	0.559

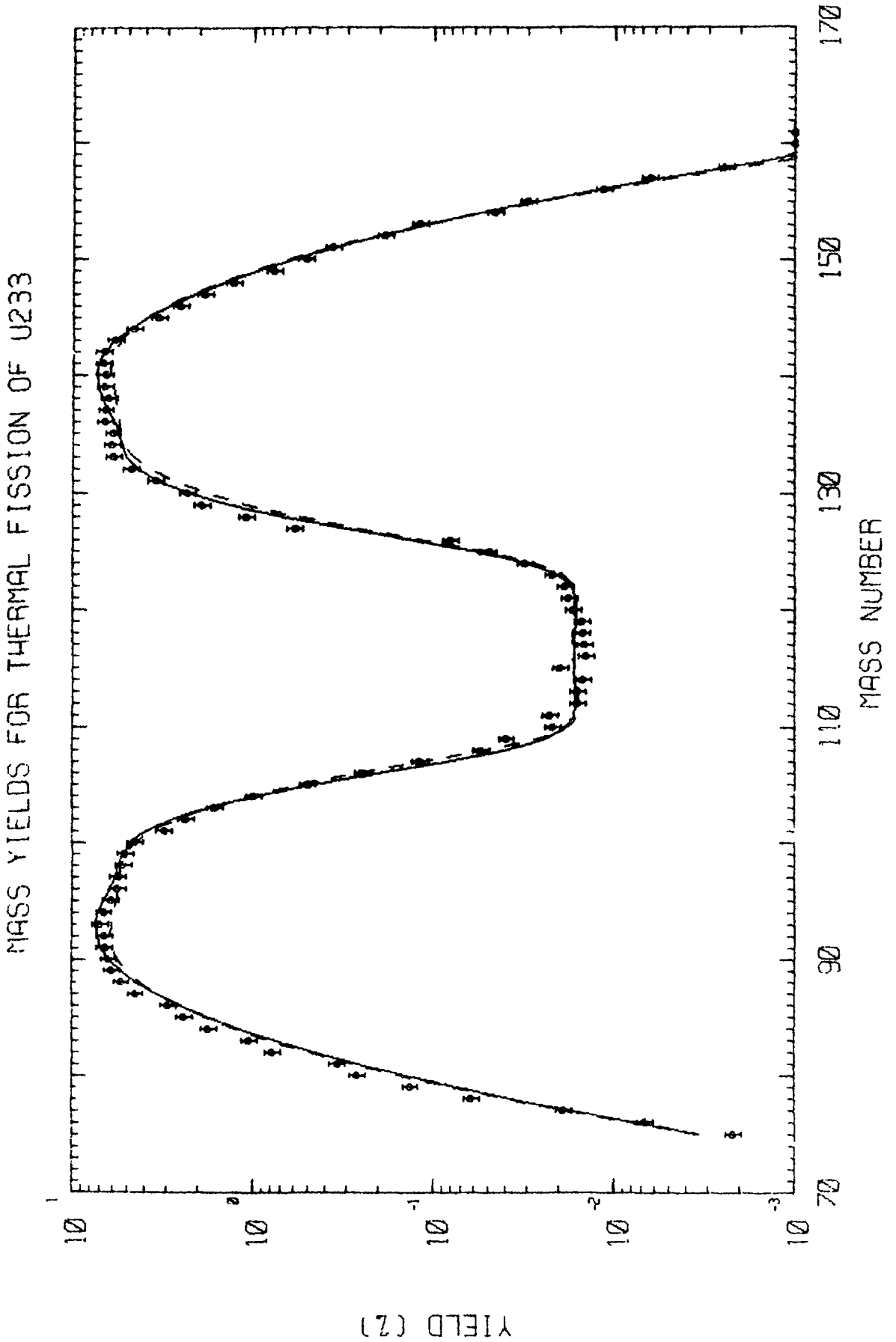


FIGURE 6a Calculated fit to $^{233}U + n_{th}$ mass yield curve with the 5 Gaussian function described in text.

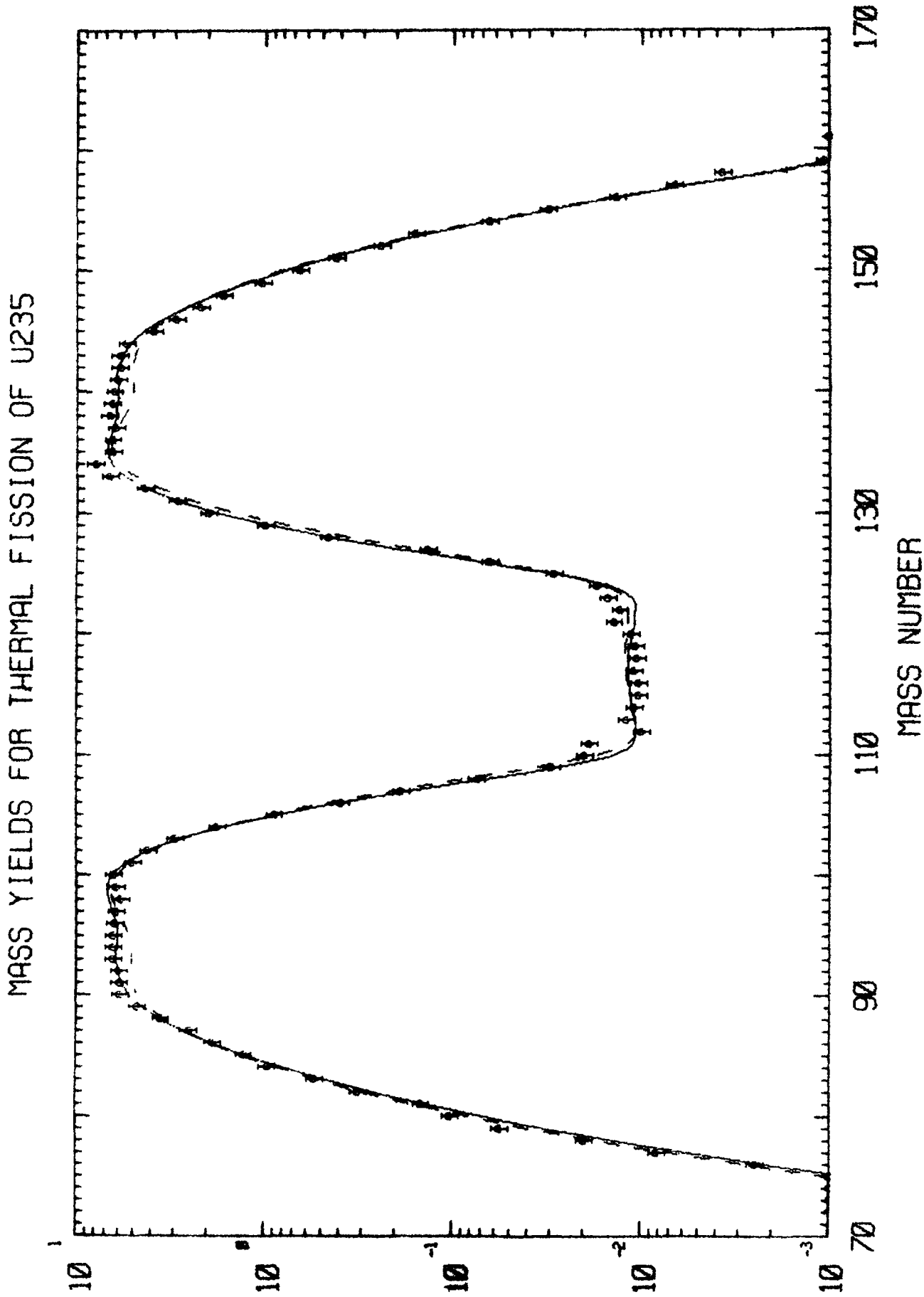


FIGURE 6b Calculated fit to $^{235}U + n_{th}$ mass yield curve with the 5 Gaussian function described in text.

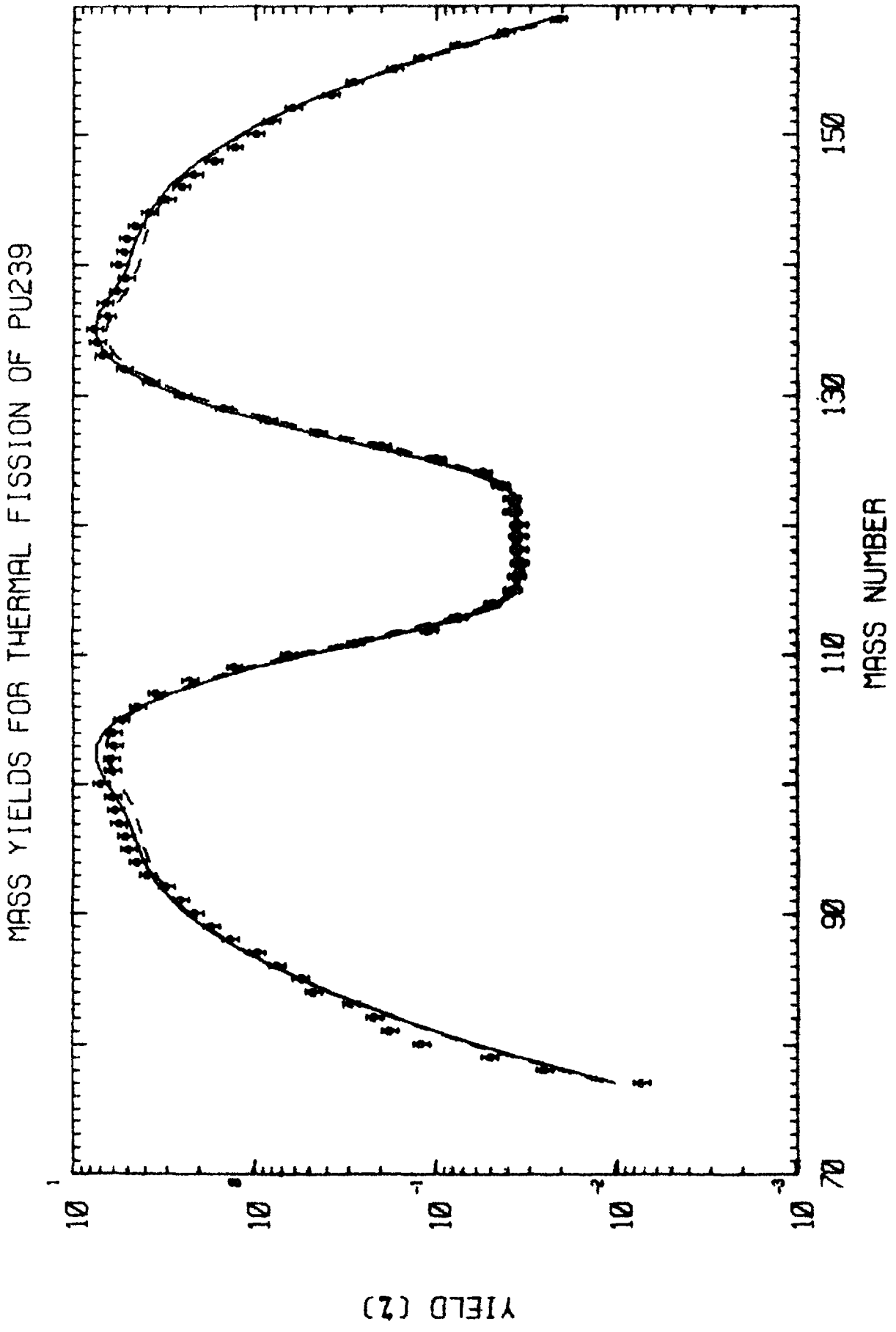


FIGURE 7a Calculated fit to $^{239}Pu + n_{th}$ mass yield curve with 5 Gaussians.

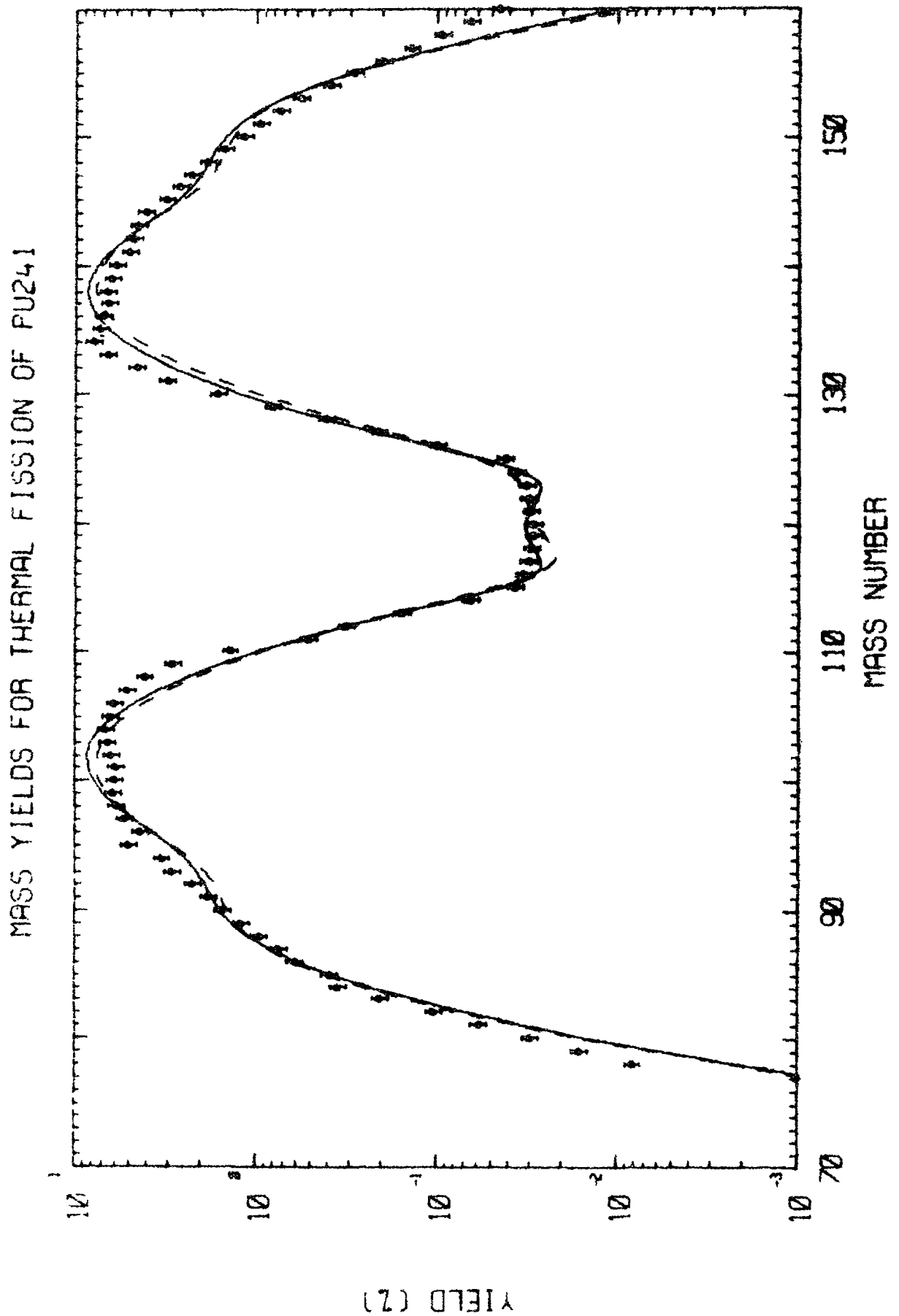


FIGURE 7b Calculated fit to $^{241}Pu + n_{th}$ mass yield curve with 5 Gaussians.

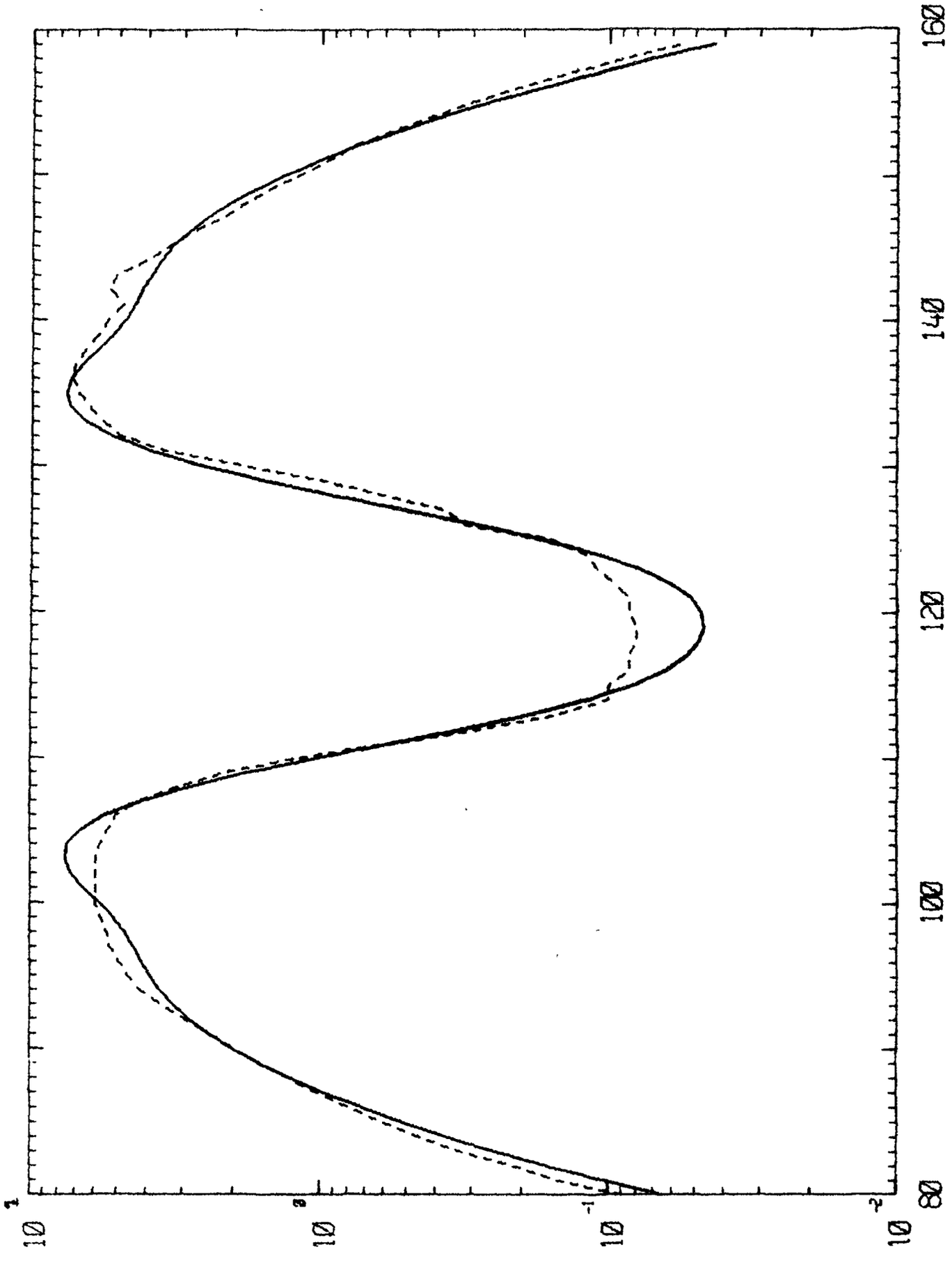


FIGURE 8 Calculated ^{240}Pu mass distribution for a fast reactor neutron spectrum using extrapolated parameters for a 5 Gaussian fit (solid line) compared with Sidebotham's [23] estimate.

TABLE II

CALCULATED INTERPOLATED YIELDS FOR ^{241}Pu

Mass No.	Ref. [19]	Ref. [21]	This work
82	0.12	0.104	0.07
89	1.20	1.2	1.3
98	5.2	5.9	5.9
100	6.2	6.05	7.9
107	6.75	6.45	6.1
108	4.0	4.15	2.9
109	2.5	2.9	1.6
110	1.2	1.4	1.0
112	0.28	0.32	0.3
114	0.07	0.065	0.08
115	0.04	0.037	0.04
116	0.03	0.033	0.03

In a final check of this method, we linearly extrapolated parameters for fit to the ^{235}U mass yield with 8 MeV neutrons [21]. The fit produced was ood and further investigation of this procedure is in progress with particular mphasis on the probable errors in the predicted yields.

CHARGE DIVISION AND DISPERSION

It has long been realised that when a fissioning nucleus divides, the harge density in the primary fragments is slightly different to that in the arent nucleus. The light fragment acquires about half a proton more and the eavy fragment correspondingly less than a proportionate share of the Z_F protons n the fissioning nucleus (see for example the review of Pappas et al. [29]. he difference between the most probable charge Z_p, for a given primary mass A nd that predicted by the postulate of unchanged charge density (UCD) is onveniently measured by the parameter $\Delta Z(A)$:

$$\Delta Z(A) = Z_p(A) - Z_{UCD}(A) \quad ,$$

here

$$Z_{UCD}(A) = \left(\frac{Z_F}{A_F}\right) \cdot A \quad .$$

Experimentally it is quite difficult to determine the quantity ΔZ for a number of reasons. Radiochemical measurement of the independent yields of selected elements has the advantage of high specificity, but most of the high yield fission fragments have very short β decay half-lives. Thus much of the data [18,30] is for the longer lived isotopes with small independent yields or for those with large fractional cumulative yields closer to the line of β stability. A most important further complication, not only confined to radiochemical measurements, is that the analysis is only accomplished after the emission of prompt neutrons from the primary fragments. In order to get back to the probable or expected primary mass which gave rise to the measured fission product mass, one must assume that the fragments with all possible primary charge splits for a given isobar gave rise, on average, to the same number of neutrons in the subsequent de-excitation process. Norenberg [31] has pointed out the dangers inherent in this assumption. However, this is forced upon us by lack of knowledge of $\bar{\nu}$ for particular fragments: only $\bar{\nu}$ averaged over groups of fragments has been measured and also now some measurements of $\bar{\nu}$ versus Z are available [32,33]. The final results are quite sensitive to this correction as can be seen for example in [29].

An alternative physical method [34,35,56] involves the separation of fission fragments of a known post neutron emission mass followed by a measurement of the average β decay chain length to stability. This method also requires a knowledge of the correction for neutrons, but even so, puzzling discrepancies between this and the radiochemical results are obtained [34].

In addition, the characteristic K X-rays emitted by the fragments can be measured in coincidence with the kinetic energies of the complementary fragments. The K X-rays identify the charge while the primary fragment mass can be worked out from considerations of energy and mass conservation [36,37]. One must assume here that the K X-ray yield is proportional to the isotopic yield for each element, an assumption which is difficult to justify [38]. Recently Cheifetz et al. [38] have reported a similar method relying not on the K X-ray but rather on the intensity of the $2^+ \longrightarrow 0^+$ ground state band transition in the de-excitation of the prompt fission product. This method is currently applicable to only even-even fragments, but avoids some of the disadvantages of the K X-ray method. Figures 9 and 10 show the results of some of the above mentioned physical methods. Note the significant difference found in the K X-ray method between values of ΔZ for different fissioning nuclei.

In early work [39,40] it was found that the then available independent yield data could be fitted with a Gaussian distribution of charge with approximately equal variance for 6 final fission product mass chains. In later work [18] with better data, a Gaussian still seemed an adequate description of the dispersion of Z values about Z_p for a given A. However, it was also clear that even Z fragments and odd Z fragments no longer seemed to be fitted exactly by the same Gaussian. There was a significant enhancement of even Z fragments. This effect was brought into even sharper focus by Amiel and Feldstein [41] who discovered that even Z yields were on average 25 per cent greater while odd Z yields were about 25 per cent less than the mean for ^{235}U thermal neutron fission.

The Gaussian charge dispersion is given by:

$$P(Z) = \frac{1}{\sqrt{2\pi\sigma^2}} \exp\left(-(Z-Z_p)^2 / 2\sigma^2\right)$$

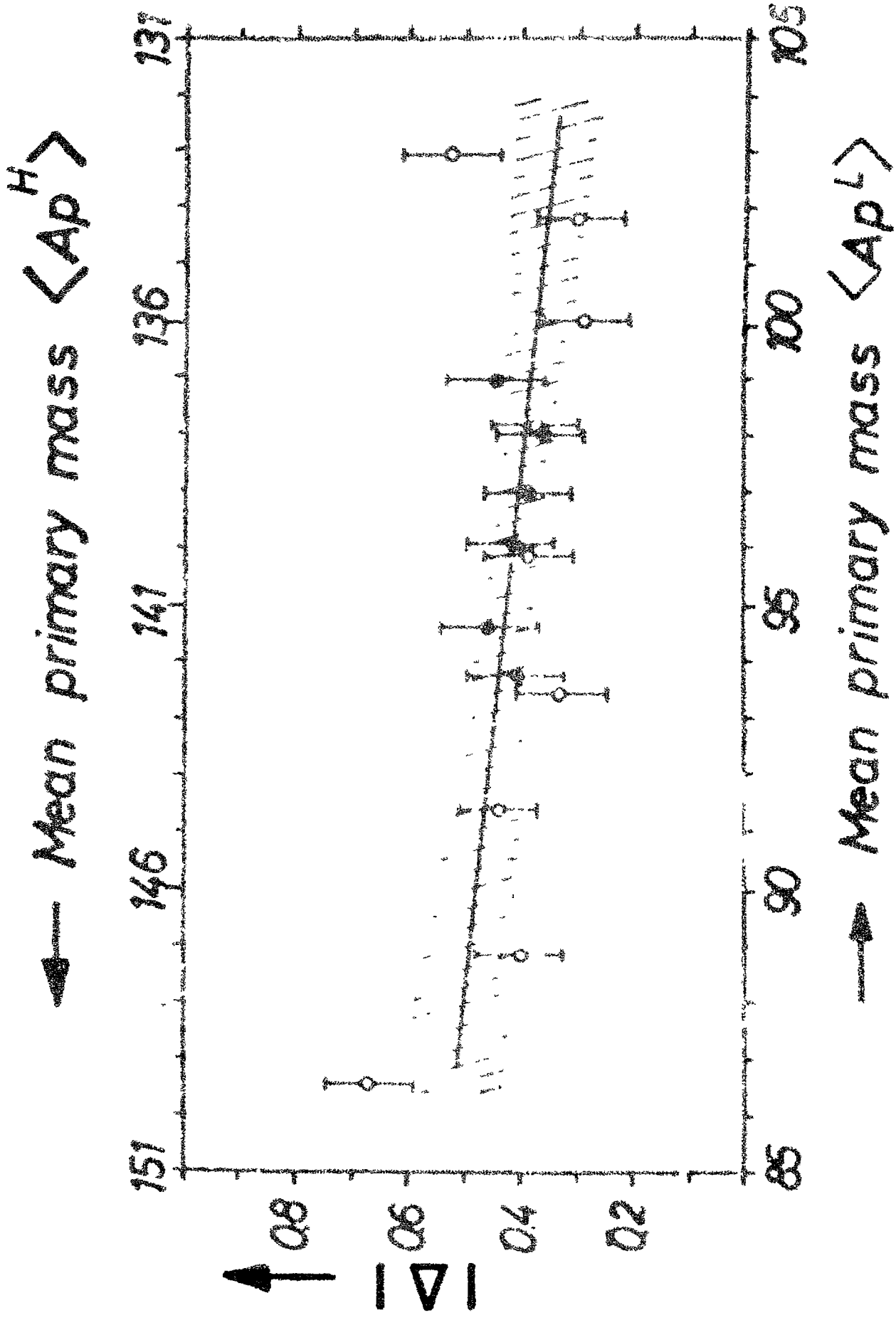


FIGURE 9 ΔZ is shown for the β-track counting method of [34]. Open and closed circles are the results for light and heavy fragments respectively. A weighted straight line fit to the data is also shown with 90% confidence levels. The post neutron emission data are corrected using an evaluated neutron emission function [34].

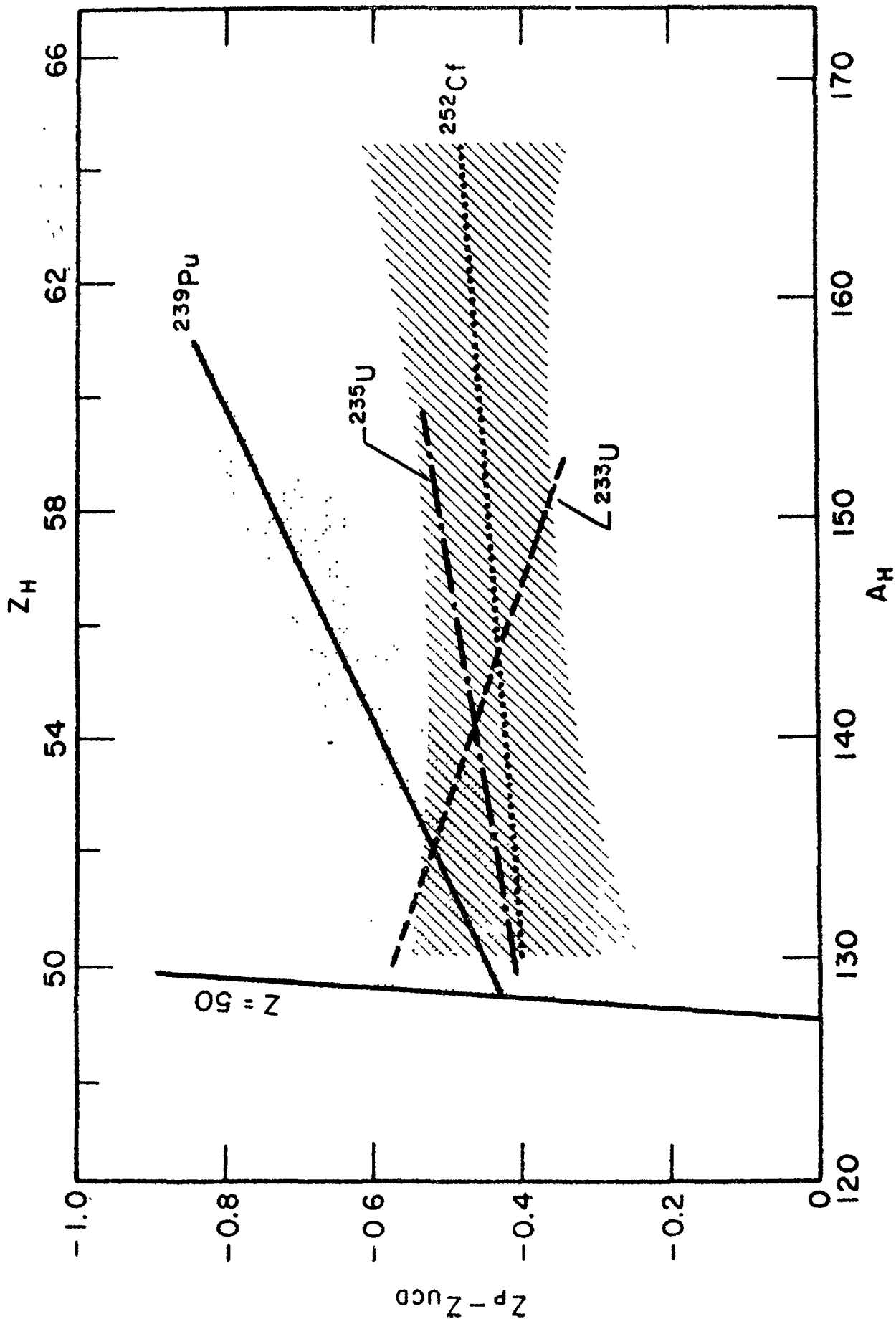


FIGURE 10 ΔZ is shown for the K X-ray method of [36] for ^{233}U, ^{235}U, ^{239}Pu fissioning with thermal neutrons and also ^{252}Cf spontaneous fission. Estimated 90% confidence areas are shown for ^{239}Pu and ^{252}Cf.

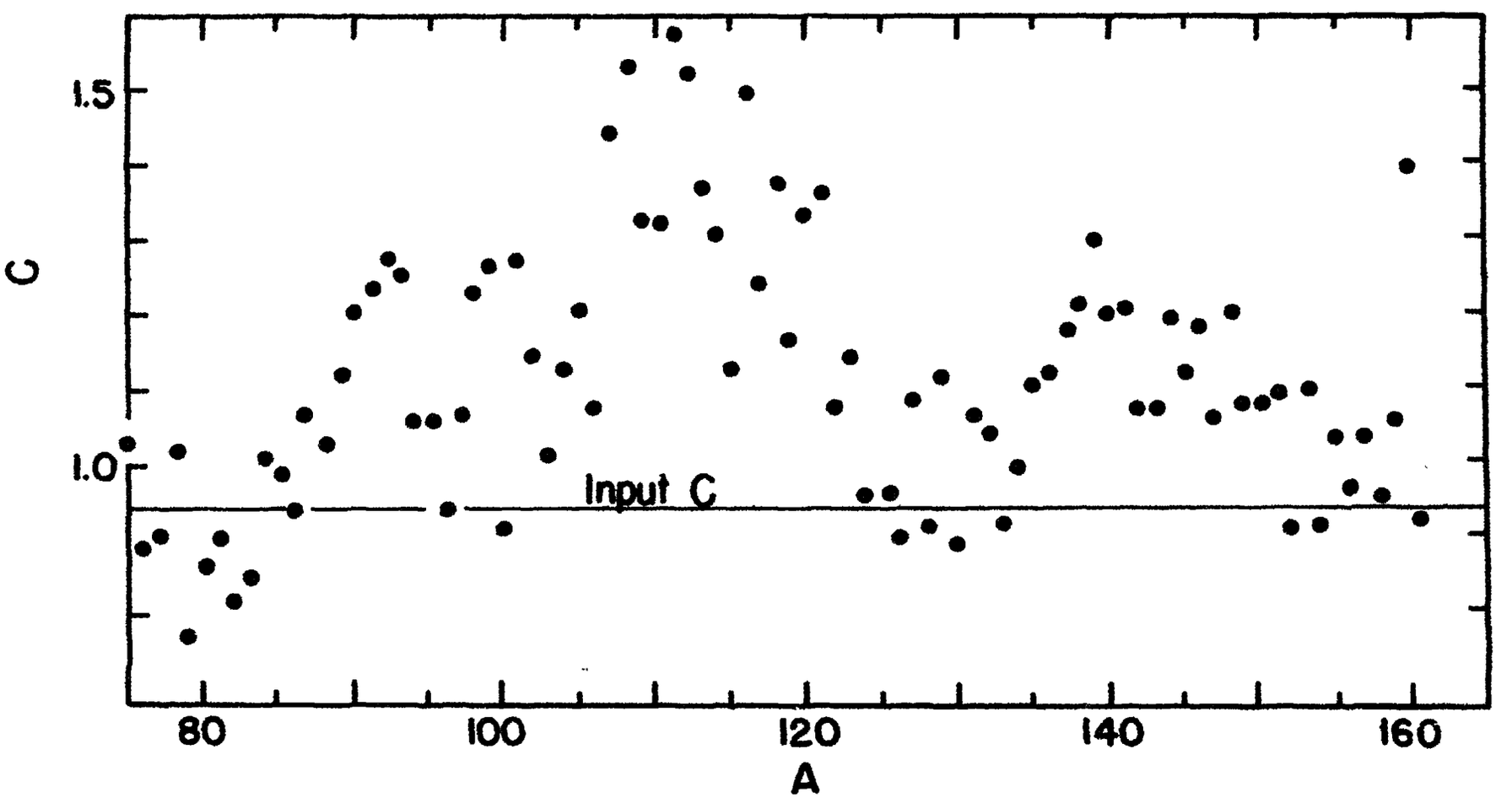


FIGURE 11 Calculated charge-dispersion parameters for the final mass yields compared with the constant input value. The charge dispersion parameter here is related to the Gaussian variance by

$$C = 2(\sigma^2 + \tfrac{1}{12})$$

and the fractional independent yield of the fission product of charge Z and mass A is

$$FIY(Z,A) = \int_{Z-\frac{1}{2}}^{Z+\frac{1}{2}} P(Z) \cdot dZ$$

Wahl et al. [18] found that most of the values of σ obtained in fitting the radiochemical data lay within a region $\sigma = 0.56 \pm 0.06$ for the high yield region away from closed shells. The data from [34] also confirm this value, but this of course reflects the situation after the emission of prompt neutrons and considerable broadening appears to be caused in the emission process [42-44]. The K X-ray method [36] previously mentioned, measured the dispersion for primary fragments of a given mass and for ^{235}U they obtained $\sigma = 0.40 \pm 0.05$. It is therefore probably fortuitious that the values of σ obtained for the post neutron emission distribution are found to be fairly constant: certainly the Monte Carlo calculations [42] show that if the initial charge distribution is constant, considerable variation would be expected in the final post neutron emission values of σ (Figure 11).

Numerous theories and semi-empirical prescriptions have been advanced to account for the behaviour of ΔZ as a function of A. The most promising theoretical approach has been to attempt to minimise for given fragment masses, the potential energy of the nascent fragments at the scission point [4, 31, 45-47]. Figure 12 shows the results obtained by Wilkins and Steinberg [4] compared with the primary charge distribution measurement of [36]. For the reasons outlined previously, it is difficult to compare the radiochemical data with theory based predictions of the pre-neutron emission charge distribution function. Any structure existing in the pre-neutron emission function could easily be removed in the subsequent neutron emissions, for example. A new evaporation model calculation is now overdue, especially since the discovery of possible considerable fine structure effects [17] in the primary mass distribution which have not been included in previous model calculations [42-44].

For most of the work in which we are interested here, a simple, clear, semi-empirical description of the ΔZ function is desired which will give reliable answers for calculated fractional yields. This can be of use in evaluations such as that of Meek and Rider [48].

Wahl et al. [18] suggest a straight line relationship for $|\Delta Z|$ for $A \geq 134$ (≤ 102):

$$|\Delta Z| = 0.45 \text{ charge units} \quad ,$$

and this is then extrapolated back to a value $|\Delta Z| = 0$ at symmetric mass division. Armbruster [46] calculates that in fact ΔZ may be 0.45 for all asymmetric fissioning nuclei, however there is some evidence that this relationship is not true for ^{252}Cf fission [49]. Mukherji [50] gives a different 'universal' empirical prescription for charge distribution, suggesting that the ratio of A/Z_p is constant for different fissioning species (where A is the primary fragment mass), but this does not appear to be substantiated by a careful examination [51].

Several authors [30, 52] have attempted to derive Z_p functions by fixing σ and fitting the yield data. Figures 13, 14 and 15 show the results obtained by Denschlag [30] for ^{235}U, ^{233}U and ^{239}Pu. Note once again the effect of different $\bar{\nu}$ versus A correction curves on the data. Alternatively, one can assume a Z_p curve and fit the Gaussian variance σ versus mass number [53]. Figure 16 shows the results and it can be seen that quite large fluctuations in σ are found. Such approaches are necessitated by the lack of sufficient data in some cases to determine both σ and Z_p simultaneously.

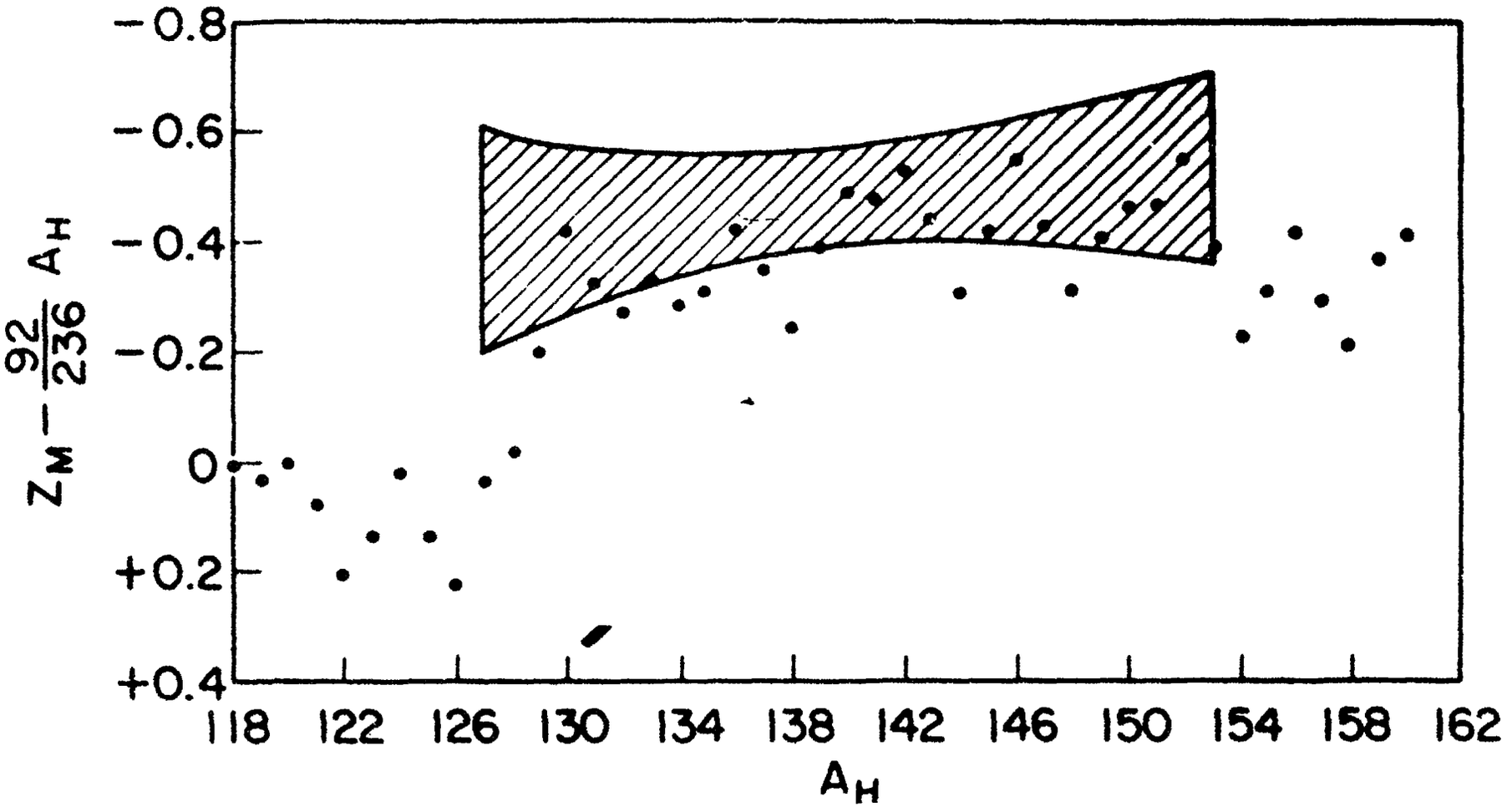


FIGURE 12 Comparison of the calculated charge distribution of ref [4] (points) with the experimental data of ref [36] for $^{235}U + n_{th}$.

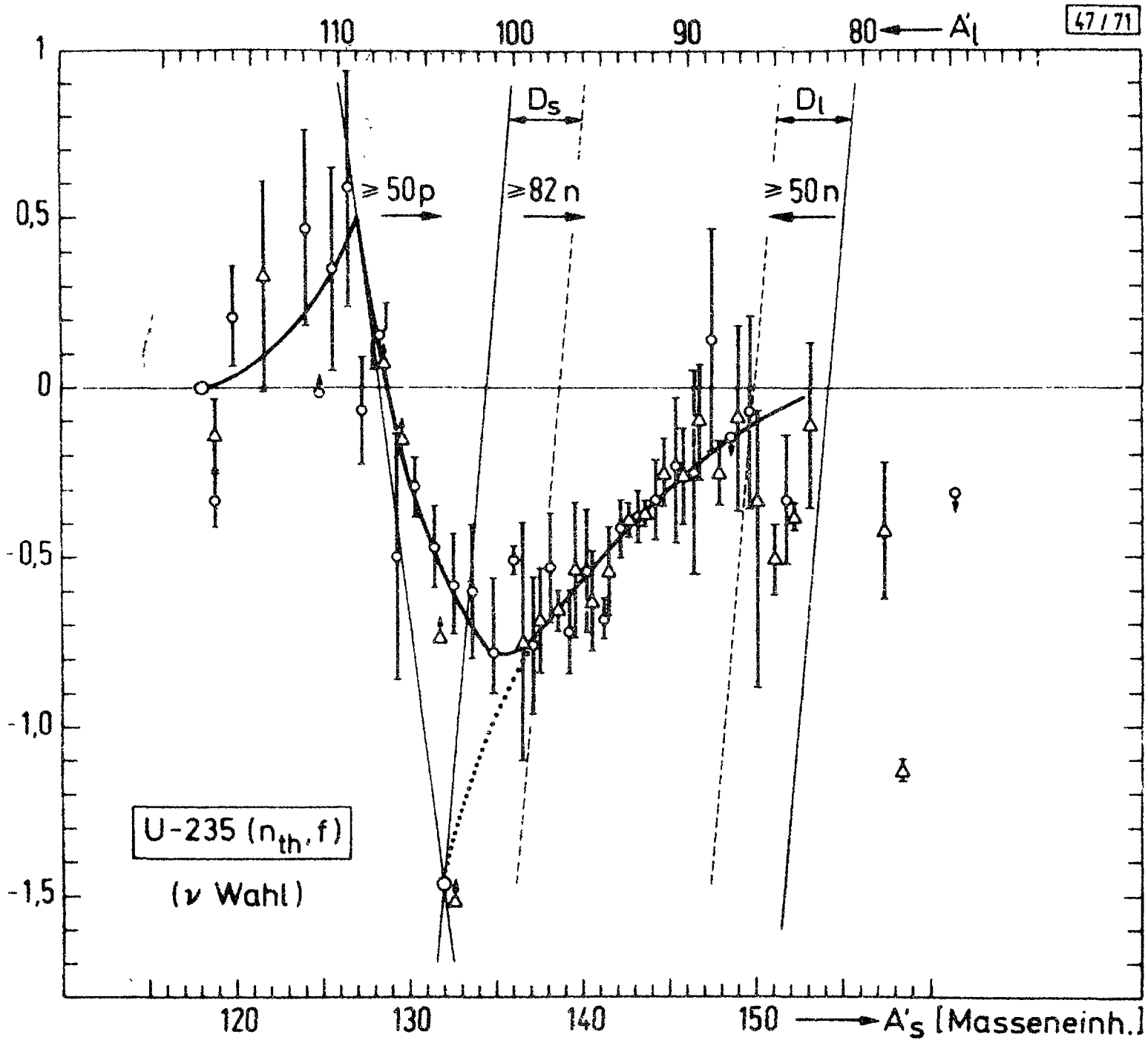
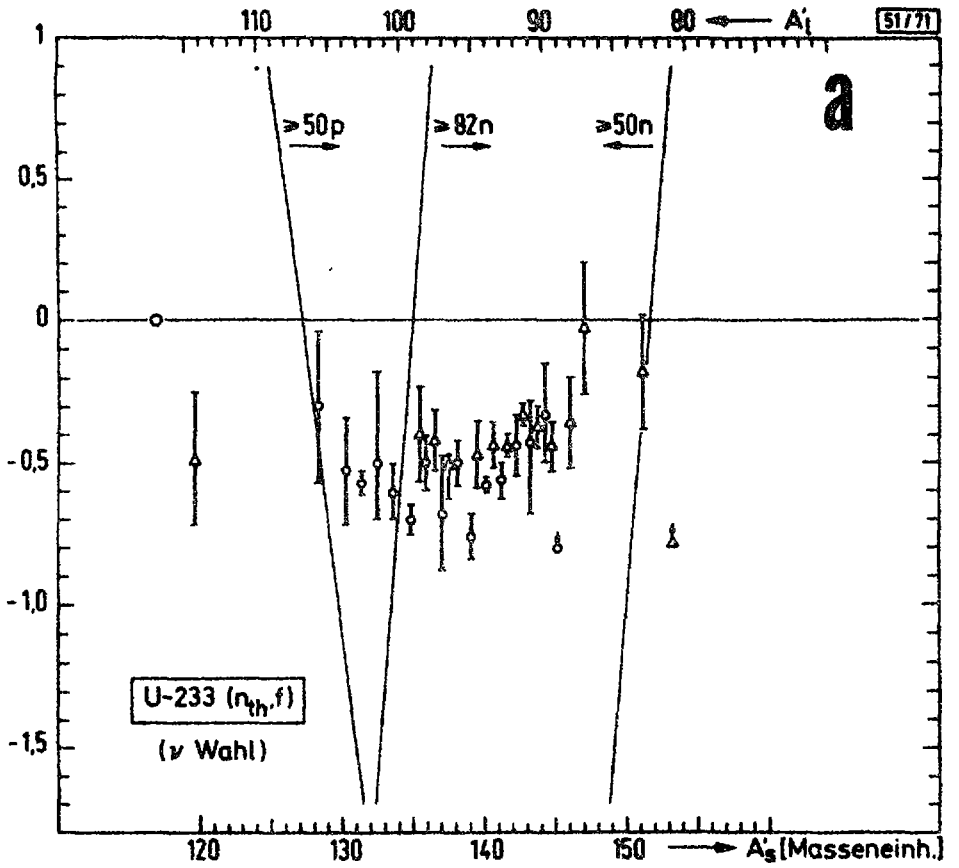
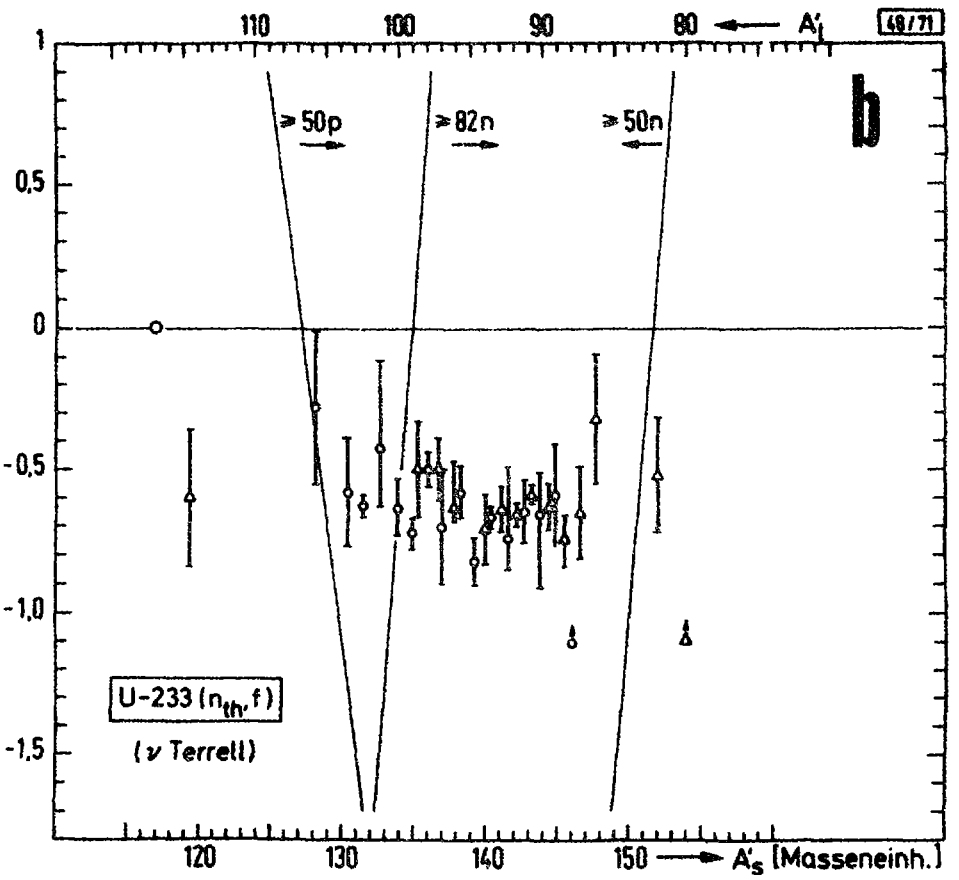


FIGURE 13 Radiochemical ΔZ for ^{235}U from Denschlag [30]. Circles are from heavy fragments while triangles indicate light fragment. A constant Gaussian width parameter $\sigma = 0.59 \pm 0.06$ was assumed for the calculation of Z_p. The neutron correction was effected using the $\bar{\nu}$ values of Wahl et al. [18]. The smooth curve shows the trend of the calculated points.





JRE 14 Radiochemical ΔZ for ^{233}U [30] showing neutron correction calculated with (a) $\bar{\nu}$ of Wahl [18] and (b) $\bar{\nu}$ of Terrell [24].

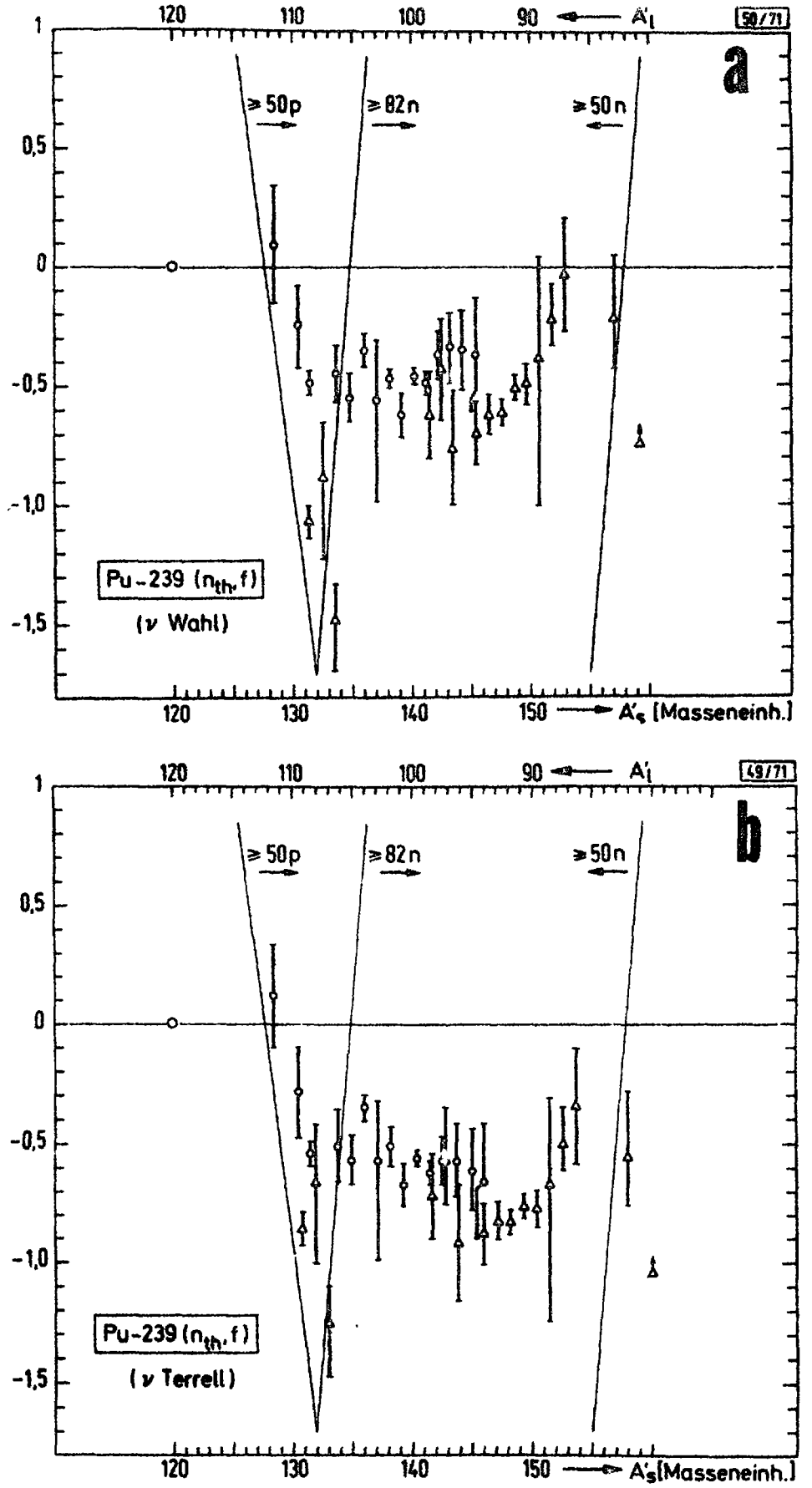


FIGURE 15 Radiochemical ΔZ for ^{239}Pu [30] showing neutron correction calculated with (a) $\bar{\nu}$ of Wahl [18] and (b) $\bar{\nu}$ of Terrell [24].

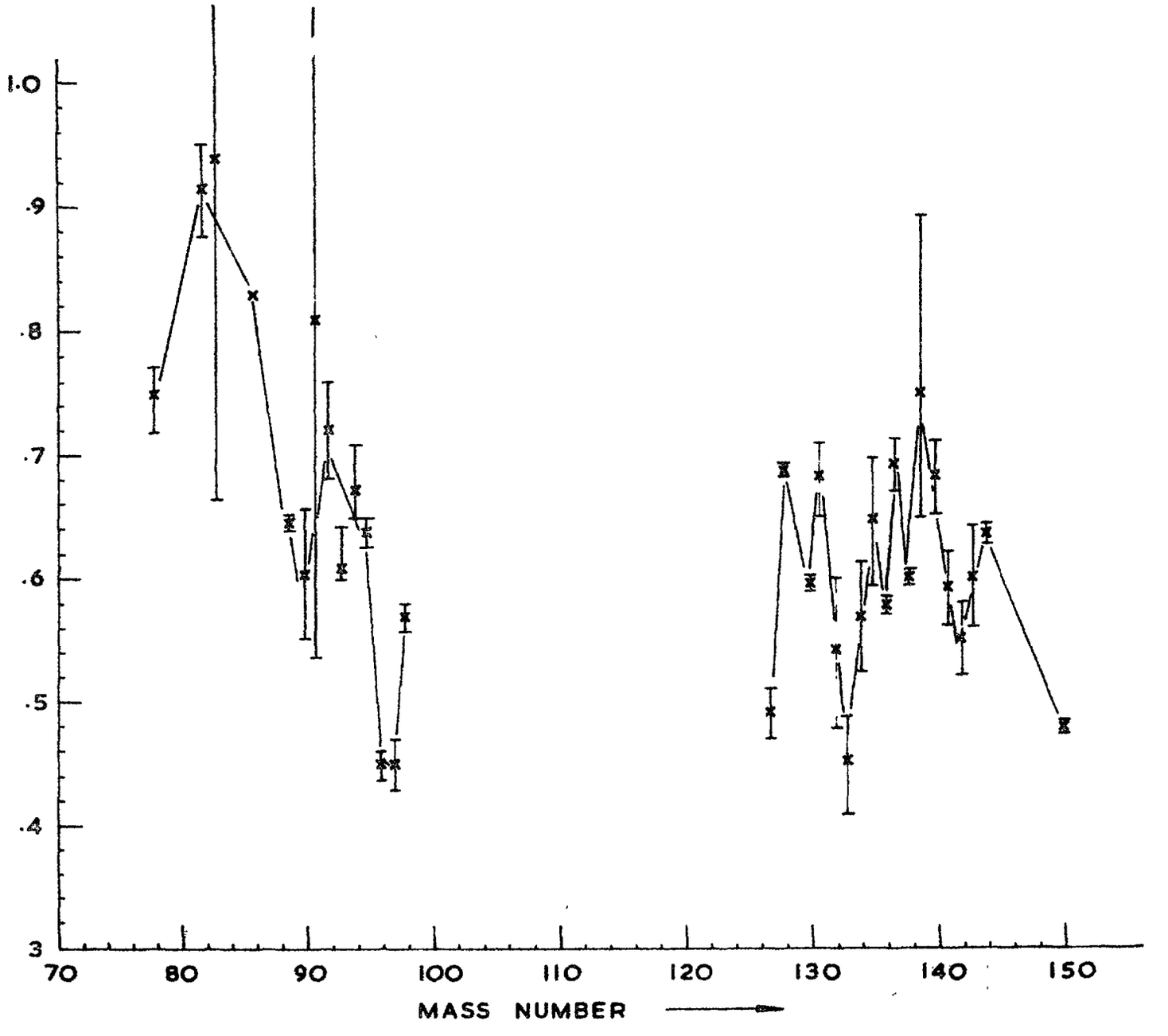


FIGURE 16 Fitted Gaussian width parameter σ versus mass number for ^{235}U thermal neutron fission [53].

We attempted for this meeting to fit all the available yield data [30] to the following functional form. The final post neutron emission charge dispersion was assumed to be described as usual by a Gaussian of constant width for all mass numbers, but with the well established (now) enhancement factor to account for even odd effects. Therefore we assumed:

$$P(Z) = \frac{1}{N(A)} \frac{(1+C)}{\sqrt{2\pi\sigma^2}} \exp\left(-(Z-Z_p)^2/2\sigma^2\right) \quad ,$$

where C is positive for Z even and negative for Z odd

N(A) is a normalisation factor to make the sum over Z for a given A equal to unity.

N(A) depends on the magnitude of C and on Z_p. We further assumed that

$$\Delta Z = -\left(\alpha + \beta\ (A-132)\right)$$

for the heavy fragments and correspondingly

$$\Delta Z = \left(\alpha + \beta\ \left((A_F-A) - 132\right)\right)$$

for the light fragments. A is the primary mass obtained by adding the $\bar{\nu}$ versus A values suggested by Wahl et al. [18] to the fission product masses. A non linear least squares fitting programme was written simultaneously to fit both the fractional independent yields and the fractional cumulative yields of [30] with the 4 parameters σ, C, α and β. The following values were obtained from a fit to the ^{235}U data:

$$\sigma = 0.569$$

$$\alpha = 0.517$$

$$\beta = -6.0 \times 10^{-3}$$

$$C = 0.206$$

Only data with $A \geq 132$ (≤ 104) were fitted to avoid the closed shell region where, since fewer neutrons are emitted, the final value of σ may be only slightly altered from the primary value $\sigma \sim 0.4$ [42] (see also Figure 11). Note that we find 20 per cent enhancement of Z even nuclei compared with the 25 per cent found by Amiel and Feldstein [41]. The values of σ and α we find are close to the values derived by Wahl et al. [18], but we would expect the addition of the parameter C to give substantially better estimates of unknown fractional independent yields. To give a rough indication of the improvement obtained in the calculated yields over that reported by Wahl et al. [18], we repeated their calculation of the yields of the complementary Sr/Xe isotopes and Rb/Cs isotopes. We obtained values much closer to the experimental ones than did Wahl et al. [18]:

	Experimental Yield	Wahl [18]	This calculation
^{37}Rb	11.9 ± 0.4%	15.1%	11.9%
^{55}Cs	11.2 ± 0.6%	15.1%	12.0%
^{38}Sr	18.6 ± 1.7%	16.0%	18.7%
^{54}Xe	19.4 ± 1.4%	16.0%	18.9%

A fit was also made to the data of ^{233}U using only Wahl's [18] values of $\bar{\nu}$. In this case the final parameters were

$$\sigma = 0.582$$

$$\alpha = 0.578$$

$$\beta = -9.0 \times 10^{-3}$$

$$C = 0.158$$

It is significant that in this case the enhancement for Z even fragments is reduced relative to ^{235}U and doubtless this is related to the fact that the system $^{233}U + n_{th}$ is excited roughly 0.5 MeV further above the fission barrier than is $^{235}U + n_{th}$ [54]. If this interpretation is correct, it points to a rather rapid decrease of the enhancement factor as excitation energy increases and this confirms the studies of fine structure versus final kinetic energy [17].

For ^{239}Pu the fit was not good, but was interesting for another reason. Again using the $\bar{\nu}$ values from [18] (note that this is the ^{235}U $\bar{\nu}$ curve) we obtained

$$\sigma = 0.67$$

$$\alpha = 0.785$$

$$\beta = -2.7 \times 10^{-2}$$

$$C = -0.087 .$$

In this case we find a negative value for C of almost 9 per cent. This does not appear to be caused by use of the wrong $\bar{\nu}$ versus A curve, but the result must at this stage be regarded as somewhat suspect until further data can clarify the situation.

A further interesting result of this work was prompted by a calculation of Reisdorf et al. [17] on the primary mass distribution. One could expect on reasonable theoretical grounds [41] that the fine structure revealed for even Z fragments in the post neutron emission case would also be present in, and to much the same extent, for even N fragments in the primary mass yields. One could (using our results) then expect an enhancement of even-even primary fragments of some 40 per cent for ^{235}U over odd-A fragments with odd-odd fragments correspondingly down. Calculations [17] show that even such large fluctuations can be masked by the mass resolution effect always present in primary mass determinations. Figure 17 shows that the final fine structure is similar to that observed experimentally for ^{235}U [12, 55].

If much of the even neutron fine structure is washed out in the emission process [18, 41] we expect the fine structure in the final mass yield curve to be closely correlated with our normalisation factor N(A) already defined, since this contains only contributions from protons. Figure 18 shows the normalisation function for ^{235}U with Walker's [19] final mass yields and it can be seen that the peaks and troughs in N(A) correspond very closely to the mass yield structure. In fact, by dividing the experimental yields by the normalising factor, the considerably smoother curve also shown in the figure is obtained. The fine structure fluctuations in the primary mass curve have to some extent been reduced although the peak at A=134 is still present. This peak is caused by the primary enhancement of N=82 which has not been fully removed, plus in this case also a secondary buildup in the neutron decay of the same magic neutron number nuclei [18]. This raises the possibility that even fine structure may be added to interpolated yield curves. Alternatively, the positions of the fine structure peaks in the final mass yield curves may be used to help determine the Z_p curve. A more detailed description of these investigations will be published elsewhere.

Since the value of the even-Z enhancement factor is expected to be quite energy dependent, it follows that the fitted value for thermal neutron fission is an average over quite a wide range of excitation energies. For low excitation energies (high kinetic energies) we would expect C to be larger than the average value and to make its effect felt on the fractional

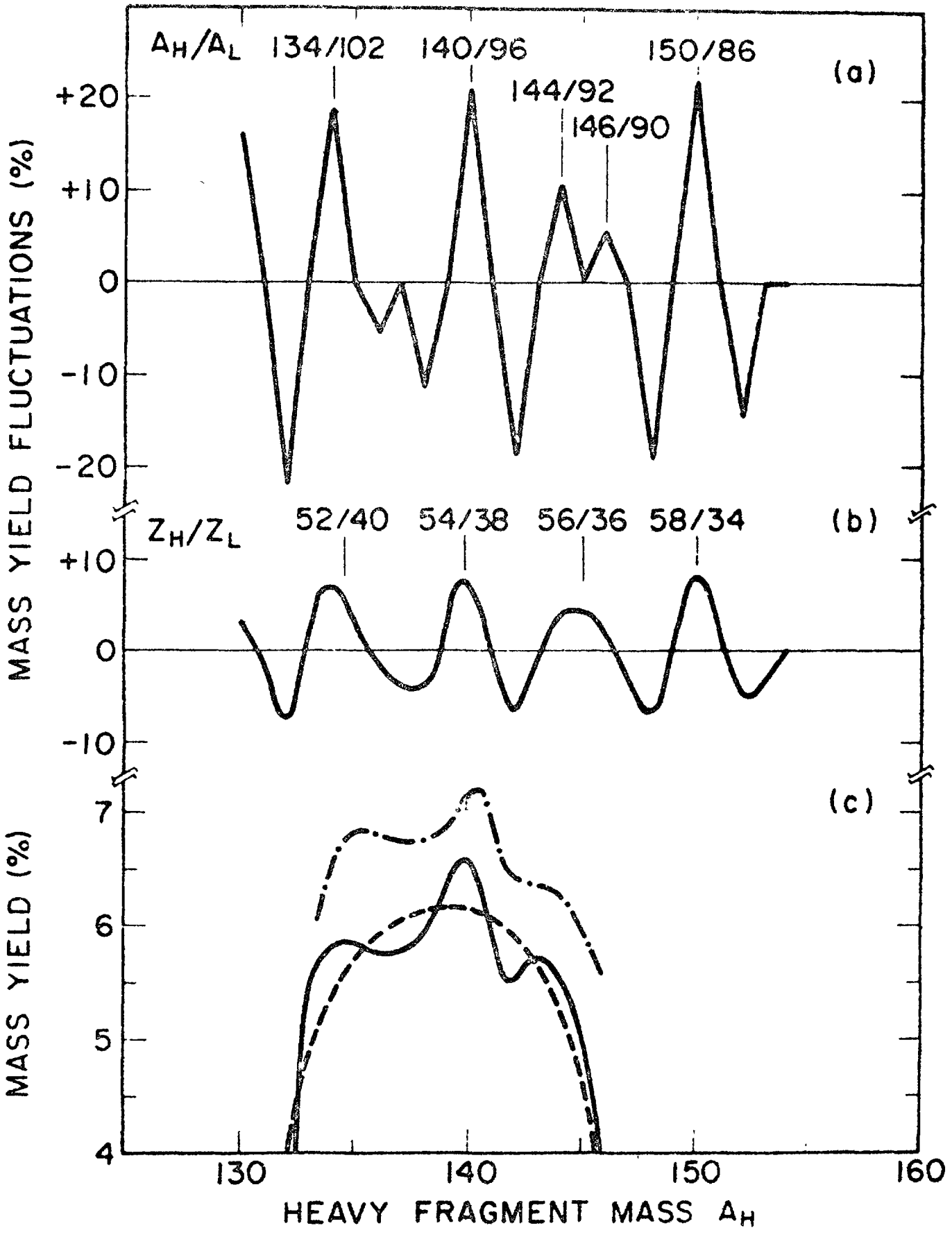


FIGURE 17 Even-Z fragment enhancement in ^{235}U. In (a) are shown the percentage mass yield fluctuations as a function of fragment mass assuming +30% enhancement for doubly even fragments and -30% for doubly odd fragments relative to the mean value for odd-A fragments. In (b) the fluctuations are shown when dispersed with a mass resolution of 2.0 amu (FWHM). The dashed curve of (c) shows an assumed smooth mass distribution while the solid curve shows the fine structure in (b) added. The dot-dashed curve in (c) is an experimentally measured mass distribution having mass resolution of ≈2 amu (FWHM).

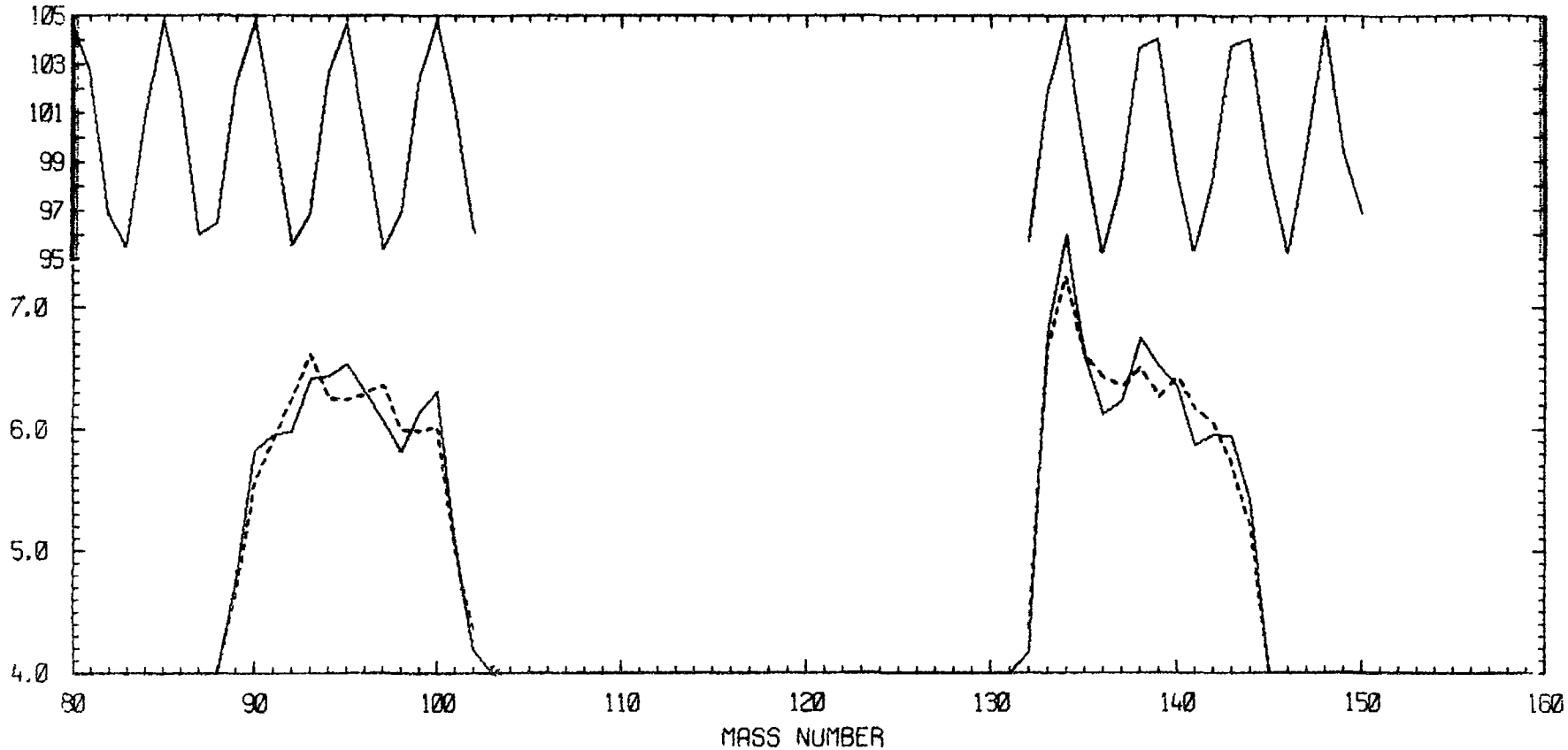


FIGURE 18 The normalisation function N(A) versus mass number is compared with the ^{235}U thermal neutron mass yield curve of [19] (solid line). The dashed curve is obtained by dividing the measured yields by the normalisation factor.

yields and mass distributions. This is confirmed by an enhanced formation probability for $^{50}Sn_{132}$ for low excitation energy events [56] since the β-counting method takes preferentially higher kinetic energy fragments than average.

We have mentioned several times the problems associated with having to correct the Z_p values to primary masses by addition of the neutron correction and we return to it here in a little more detail. Nubar versus A for ^{235}U has been measured experimentally by several authors [56-60] but the measurements are often in disagreement by more than the quoted errors. Sistemich et al. [34] and also Wahl et al. [18] have produced evaluated $\bar{\nu}$ versus A curves, but although Sistemich et al. [34] renormalise the data of reference [58] neither in [34] nor in [18] is it apparently realised that the data of Milton and Fraser [58] have not been corrected for neutrons emitted backwards from the complementary fragment. This correction can be quite significant as before this correction was applied the data of [60] were in good agreement with that of [58] while after the correction they are in good agreement with the data of [59]. Thus if the data from [57] are disregarded as they do not agree with the $\bar{\nu}_T$ values obtained from the reliably known secondary mass yields [18], the remainder are probably in substantial agreement.

There is a further complication which arises from the fact that measured $\bar{\nu}$ versus A curves refer to the primary (pre-neutron emission) mass and what is required to correct the post neutron emission Z_p values is $\bar{\nu}$ versus the post neutron emission mass. The evaluation of Wahl et al. [18] appears to be the only one specifically referring $\bar{\nu}$ to the final mass number. In principle, the method used by Wahl et al. [18] should be fairly reliable since it uses $\bar{\nu}_T$ calculated by the method of Terrell [24], combined with the ratio ν_L/ν_H estimated from experiments [57-60]. However, even apart from Wahl's use of data from [58] to estimate ν_L/ν_H, there is a further correction required for mass resolution [24]. This will cause the slope of the $\bar{\nu}$ versus A curve to alter slightly. Therefore we have made a new evaluation of $\bar{\nu}$ for ^{235}U. The values of $\bar{\nu}_T$ were found using Walker's [19] yield data and assuming $\bar{\nu}$ at symmetry to be 4.0 [18]. The ratio of ν_L/ν_H was taken from the data of [59,60] only and a rough correction was made for mass resolution. Remembering that the ν_L/ν_H ratio referred to the primary mass, we calculated the average number of prompt neutrons ν_p emitted in conjunction with the final mass A. These are given in Table III. Also, not ready in time for this review are new evaluations of the $\bar{\nu}$ data for ^{233}U and ^{239}Pu. There are some significant differences between the $\bar{\nu}$ values of [18] and those given here. Between mass numbers 80 to 102 the new values are on average 0.15 neutrons less than the old, while the large peak near mass number 110 has now been removed since this does not appear in the data of [59] or [60]. The new values for the heavy fragment masses are little changed over the old ones. The calculated $\bar{\nu}_T$ for our data is 2.40 in good agreement with experiment.

If a new evaluation of $\bar{\nu}$ versus A was necessary for ^{235}U, it is even more desirable for ^{233}U and ^{239}Pu since there does not appear to have been an attempt to produce such an evaluation since 1962 [24]. Until this is done, it is difficult to compare Z_p curves from different fissioning nuclei. The work of Gunther et al. [56] on β-track counting appears to indicate that Z_p may be the same for the heavy fragment post neutron emission masses of ^{235}U and ^{239}Pu. This result, since it refers to the post neutron emission situation, is not dependent on knowledge of a $\bar{\nu}$ versus A curve for each nuclide. There appears to be some experimental difficulties in this work [62], but if this result were generally true, then $|\Delta Z|$ would be different for each fissioning nuclide. Our own fitting procedure has also uncovered some differences in $|\Delta Z|$ from the three cases studied (admittedly using $\bar{\nu}$ curves likely to be considerably in error for ^{233}U and ^{239}Pu).

TABLE III

FINAL ν_T AND CALCULATED FRAGMENT NEUTRON NUMBERS VERSUS MASS NUMBER (POST NEUTRON EMISSION)

A	ν_T	ν	A	ν_T	ν
77	2.50	0.69	101	1.92	1.25
78	2.50	0.69	102	1.87	1.27
79	2.50	0.70	103	1.90	1.35
80	2.50	0.71	104	1.91	1.42
81	2.50	0.72	105	1.90	1.48
82	2.50	0.73	106	1.87	1.47
83	2.55	0.76	107	1.93	1.49
84	2.58	0.80	108	2.50	1.87
85	2.55	0.83	109	2.85	2.02
86	2.53	0.87	110	3.23	2.03
87	2.52	0.93	111	3.62	2.14
88	2.54	1.02	112	3.69	2.13
89	2.54	1.10	113	3.91	2.12
90	2.54	1.17	114	4.00	2.09
91	2.54	1.22	115	4.00	2.03
92	2.56	1.28	116	4.00	2.00
93	2.60	1.34	117	4.00	1.97
94	2.59	1.37	118	4.00	1.91
95	2.57	1.40	119	3.89	1.80
96	2.55	1.41	120	3.83	1.69
97	2.53	1.43	121	3.64	1.52
98	2.44	1.41	122	3.44	1.37
99	2.32	1.40	123	3.14	1.14
100	2.17	1.36	124	2.86	0.87

TABLE III (cont'd)

A	ν_T	ν	A	ν_T	ν
125	2.57	0.69	141	2.59	1.28
126	2.23	0.56	142	2.55	1.30
127	1.99	0.45	143	2.54	1.36
128	1.72	0.37	144	2.56	1.42
129	1.76	0.37	145	2.56	1.49
130	1.83	0.45	146	2.55	1.56
131	1.84	0.51	147	2.54	1.63
132	1.82	0.56	148	2.55	1.69
133	1.87	0.64	149	2.60	1.77
134	2.18	0.82	150	2.56	1.78
135	2.40	0.97	151	2.55	1.79
136	2.50	1.07	152	2.50	1.79
137	2.53	1.11	153	2.52	1.80
138	2.54	1.15	154	2.50	1.80
139	2.59	1.20	155	2.50	1.81
140	2.60	1.25	156	2.50	1.82

In summary, for the purposes of calculating unmeasured fission product yields it is possible to use some semi-empirical function fitted to the post neutron emission fractional yield data. The function fitted to the data of ^{235}U may not be sufficiently 'universal' for use in other fissioning systems but no doubt it will continue to be used in lieu of any alternative.

Finally, it now appears desirable to perform new evaporation model calculations with a view to investigating more fully than has been previously possible, the relation of post neutron emission yields to the primary mass yields. The effects of fine structure would be of great interest and has not been included in previous calculations [42-44]. It seems likely that if the primary mass distribution has significant fine structure then so will $\bar{\nu}$ versus A, Figure 19, [61] and possibly also Z_p as a function of A. These questions are perhaps not so pressingly important for our task of prediction of unmeasured fission product yields as they are for a fuller understanding of the fission process.

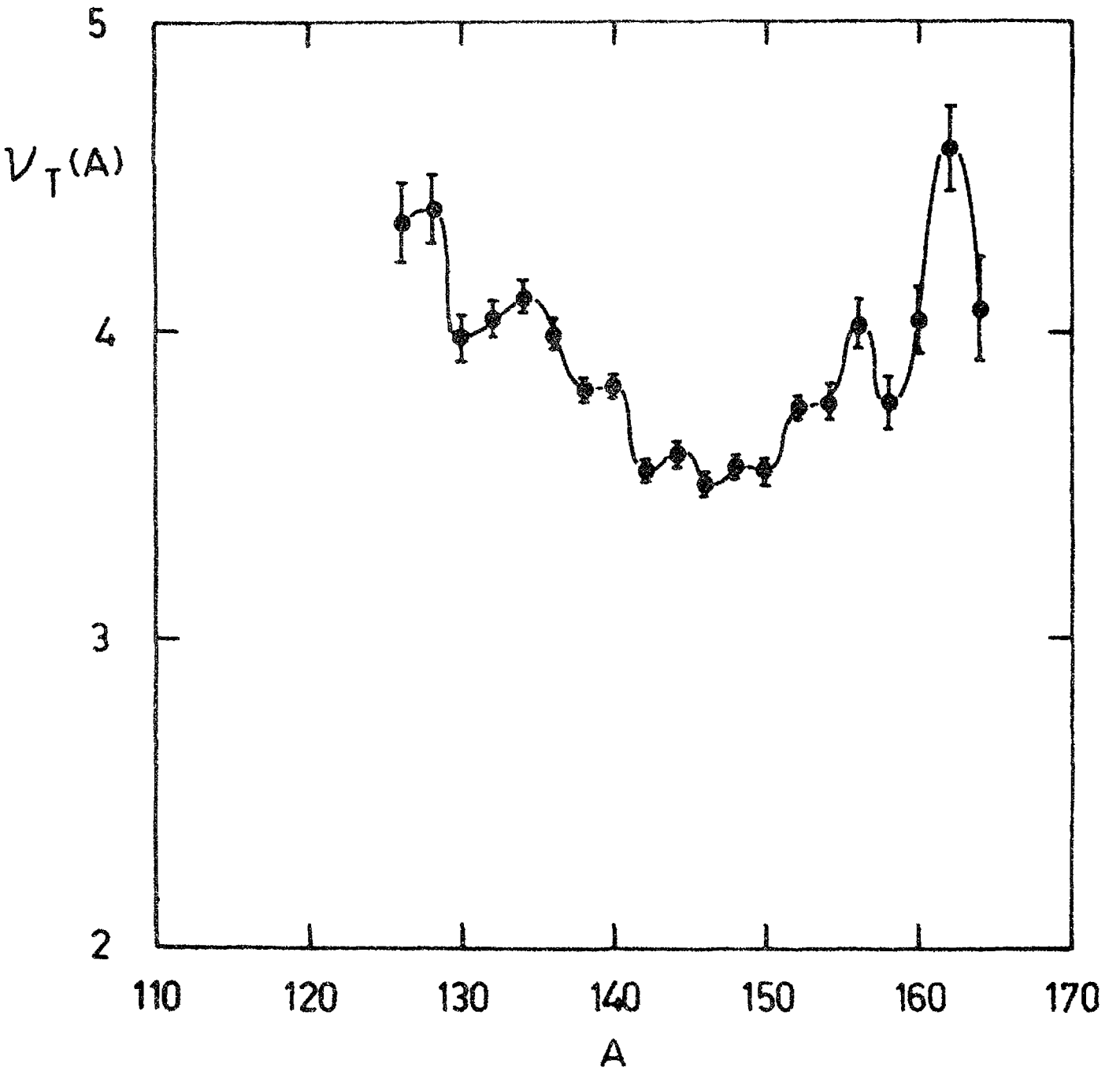


FIGURE 19 Recent data on $\bar{\nu}_T$ versus A for ^{252}Cf [61] showing fine structure peaks. The peaks are correlated with the fine structure peaks in the mass distribution.

5. REFERENCES

The following conferences are referred to below in abbreviated form:

Salzburg Conference (1965): IAEA Symposium on Physics and Chemistry of Fission, IAEA Salzburg, Austria (1965).

Vienna Conference (1969): Second IAEA Symposium on Physics and Chemistry of Fission, IAEA Vienna (1969).

Rochester Conference (1973): Third IAEA Symposium on Physics and Chemistry of Fission (not yet published).

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S u m m a r y

OBSERVATIONS, CONCLUSIONS AND RECOMMENDATIONS OF THE PANEL

In view of the complexity of the topics discussed at this Panel meeting, the observations, conclusions and recommendations on different major subjects are grouped together in the following seven chapters:

Chapter 1: Introduction (General observations, conclusions and recommendations).

Chapter 2: International cooperation in the exchange and dissemination of FPND information.

Chapter 3: FP inventory and decay heat.

Chapter 4: FP yield data.

Chapter 5: FP decay data.

Chapter 6: Delayed neutron data.

Chapter 7: Neutron cross-sections.

A summary of the important recommendations is given in Chapter 8. Comparisons of user requirements and data status for individual FPND are presented in Appendices A1 - A5:

Appendix A1: FP chain yields.

Appendix A2: Independent and cumulative fission yields.

Appendix A3: FP decay data.

Appendix A4: Neutron reaction cross-sections.

Appendix A5: FP decay heat.

1. INTRODUCTION

1.1 Scope

The Panel achieved a first review of requirements, status and availability of fission product nuclear data (FPND) important for various fields of practical applications. The topics discussed by the meeting are reflected by the titles of the review papers. The scope of the Panel was limited to these topics, as discussed in Review Paper 1a (but see also 1.2.1-(iv)).

The FPND considered by the Panel were of the following categories:

A. Yields (cumulative and independent)

B. Decay data

C. Delayed neutron data

D. Neutron reaction cross-sections

These categories included also integral FPND such as the total decay energy released after reactor shutdown (B) and total absorption of lumped FP (D).

This was the first meeting where users and producers of FPND met to compare required FPND accuracies with the status of available FPND and to discuss further experimental and evaluation work needed and measures for an improved communication between FPND users and producers Apart from their own experience, the Panel participants relied on the background information supplied by the review papers. These review papers were internationally coordinated incorporating contributions from many experts in the field in order to provide a broad spectrum of opinions and to include also most recent experimental results.

The observations, conclusions and recommendations issued by the Panel are intended to stimulate coordinated activities in various laboratories whose results should be reviewed in a follow-up meeting of the same kind.

1.2 General observations and conclusions

1.2.1 User requirements

(i) The FPND and their accuracies required by users as observed by the Panel are discussed in chapters 2-7 and summarized in the Appendices A1-A5, together with the status of required FPND. These

appendices represent a first broad picture of the present knowledge of FPND requirements.

The Panel noted, however, that, with some exceptions, the requirements were not sufficiently supported by sensitivity studies relating requested FPND accuracies to those needed for the prediction of relevant technological parameters, and that a more thorough assessment based on the available experimental evidence was generally also lacking. It also noted that other users not present at this meeting might have different requirements.

Therefore the Panel wishes to emphasize the preliminary nature of the presently compiled requirements, but expresses the hope that they will stimulate critical comments and more detailed investigations and thus help to pave the way towards a better screened true "international FPND request list" (see recommendation in chapter 2).

(ii) The discussions on FPND requirements were generally limited to applications already in use. While the Panel recognized the importance of experimental studies of specific problems and the development of new methods in application fields, not all of these topics could possibly be covered at this meeting.

(iii) Review paper no. 6 discusses in detail the role of FPND in nuclear materials safeguards. In summary, the methods in safeguards that need FPND are not used routinely but only in special cases. These methods, although already used in test cases, need further development and detailed investigations of their applicability. Therefore FPND requirements for safeguards have low priority compared to other user needs and further sensitivity studies are necessary (see (i) above).

(iv) The scope of the Panel was limited to the discussion topics of the review papers. However, the Panel considered three more topics to be worth discussing during the meeting:

Photoneutrons are of importance, especially in reactors containing heavy water or Be. A significant number of photoneutrons is produced in these reactors from high energy bremsstrahlung and γ-rays produced in FP decay, as well as from fission and capture γ-rays. This topic was discussed only briefly and the individual FP's in question have not yet been identified, but general statements on the relevant FPND are included in chapters 4 and 5.

- Fuel element design was briefly discussed during the meeting. R.H. Flowers, an expert in this field, supplied the Panel with background information and data requirements, which are reproduced in chapter 3.

- In fast reactor dosimetry fission yields are required for measurements using ^{232}Th, ^{235}U, ^{236}U, ^{238}U, ^{237}Np and ^{239}Pu as fluence monitors. Needs for the LMFBR and FTR programme at Hanford (USA) were presented to the Panel by R.E. Schenter and are included in Appendix A1, Table A1-III. U. Farinelli summarized the conclusions of the IAEA Consultants Meeting on Nuclear Data for Reactor Neutron Dosimetry, held in Vienna from 10-12 September 1973. ("Others" in Appendix A1, Table A1-III).

1.2.2 Status of FPND

Reviewers of the status of FPND had the task of surveying existing evaluations supplemented by recent experimental results. The Panel noted that the assignment of uncertainties by evaluators was not always satisfactory, in some cases even missing, particularly for FP decay data.

On the basis of the Panel discussions FPND uncertainty figures were compiled from selected evaluations. They are listed in Appendices A1-A5 to allow a comparison with user requirements. The Panel discussed and suggested improvements of evaluations in general which are outlined in section 1.2.3. Special requirements pertaining to evaluations of specific FPND are included in chapters 4 to 7.

1.2.3 Evaluation

First the Panel recalled that the essential task of the evaluators is to critically review available experimental data and provide users with a set of "best" values. However, in order to enable the user to judge the quality of an evaluation and rely on the values recommended, it is important that the evaluator documents in detail the experimental data basis, the method and the results of his work. The presently available evaluations of FPND fall partly short of this ideal.

The experimental data considered in the evaluation, the physical conditions under which they were obtained, sources of statistical and systematic errors and discrepancies between the data are often not documented and probably not satisfactorily investigated; the Panel noted, however, a few exceptions where evaluators had been able to correct original data and thus

resolve discrepancies between them. Furthermore FPND evaluators sometimes do not assign properly assessed uncertainties to their recommended data (and sometimes cannot do so because of the inadequacy of the experimental data); such uncertainties are often very important to the user and will influence his choice of an evaluation. In this context the Panel observed that evaluators, in order to cope satisfactorily with these tasks, should be able to judge experimental results adequately. This would be easier if the experimenters would describe their measurements in sufficient detail and, in particular, perform a satisfactory error analysis separating random and systematic errors.

The above mentioned shortcomings are some of the reasons why different evaluations of FPND usually show different results. Other reasons consist in differences in the experimental data basis available to the evaluator, in unresolved discrepancies between different experimental data and justifiable differences in the evaluators' judgement and methods of analysis of the experimental data. Also the objective of an evaluation and the time and effort an evaluator is allowed to spend on an individual task have an influence on the final result and its quality.

These observations led the Panel to express concern not to aim at the establishment of only one standard FPND library, which some users would find convenient, but to pursue several independent evaluation efforts: The users should not have to rely on the results of one evaluation only, but be able to choose among different evaluations and to select the one best fitting their purposes. The adoption of a standard format for data, as discussed later, is a key step in making this approach workable.

1.2.4. FPND user-producer communication

It was the impression of the Panel that the simultaneous presence of measurers, evaluators and users of FPND at one meeting stimulated fruitful discussions resulting in a better understanding of each others' problems. Closer contacts in the future should be established by improved ways of communication as suggested by the panel and outlined in chapter 2.

1.3. General Recommendations

(i) The Panel recommends that users of FPND perform sensitivity studies to enable a better specification of their requirements.

(ii) FPND evaluation work should continue to be performed at different places.

(iii) Evaluators are requested to publish all pertinent details of their work. They should attempt to identify systematic errors and resolve discrepancies. They should assess random and systematic errors separately, assign uncertainties to their recommended data and warn users in cases of unresolved discrepancies. Recognizing the magnitude of the work which is implied in these requirements and its importance to FPND users the Panel recommends that in future stronger support be given to FPND compilation and evaluation.

(iv) Measurers are requested to publish all details on experimental conditions, corrections applied and error analysis required for an adequate comparison with other measurements. A recommendation to improve the intercommunication between measurers and evaluators in this matter is included in chapter 2.

(v) A follow-up panel should be convened in about three years to review the progress in FPND measurement and evaluation, and sensitivity studies stimulated by the present meeting. Discussion topics should again be covered by review papers in order to provide background and save time for discussion. It is to be hoped that this follow-up meeting will be in a position to set up a final list of user requirements based on sensitivity studies.

(vi) Surveys of user requirements of FPND should be completed and distributed to reviewers of the status of FPND well in advance of the follow-up panel, so as to give the status reviewers sufficient time to prepare lists of uncertainties for the required FPND.

2. INTERNATIONAL COOPERATION IN THE EXCHANGE AND DISSEMINATION OF FPND INFORMATION

2.1. Observations and conclusions

(i) The Panel noted in general that a regular exchange and dissemination of information in the field of FPND is lacking.

(ii) The list of FPND compilations and evaluations provided by Valente from NEA/CCDN (review paper 1b) to the meeting was found most valuable by the Panel participants.

(iii) The Panel observed that, in spite of the large number of existing FPND compilations listed in Valente's review paper, evaluated FPND are only partially included in the most widely used computer files of evaluated nuclear data.

(iv) The recent inclusion of FP yield and decay data in the US ENDF/B library was considered a great step forward. FPND data types and associated physical quantities foreseen in the ENDF format are specified in Annex 1 to this chapter. Some critical remarks concerning these specifications are given in Annex 3.

(v) One of the main difficulties encountered in the comparison and mutual conversion of different evaluated nuclear data files are the differences in physical content associated with certain classes of data. Such differences should be avoided when new classes of data are introduced and should, where possible, be eliminated for data already existing in files.

(vi) During the meeting it was frequently observed that the communication between the measurers, evaluators and users of FPND is still unsatisfactory:

- sources of available evaluated FPND are not sufficiently well known to users;
- users and evaluators have no means to inform FPND measurers about their requirements, except in the field of neutron induced reaction data, where WRENDA exists.
- the communication channels between FPND measurers, evaluators and users via presently existing publishing media are too slow, thus affecting also the efficient planning and coordination of experimental and evaluation work;
- at present there exist no convenient means of informing those interested in FPND about observed discrepancies.

2.2. Recommendations

(i) The Panel recommends that the list of FPND compilations and evaluations as provided by Valente from NEA/CCDN to this meeting be kept up-to-date and published at annual intervals.

In order to enhance the value of the list, it is recommended that future issues should contain short comments by the authors to each reference concerning its content, up-to-dateness, application area and a specification of the availability in computer medium of the data concerned including the computer format.

The first updated list should be published not later than one year after the panel. The Panel participants leave it to the discretion of NEA/CCDN, IAEA/NDS and other nuclear data centres to decide which centre will publish the list in the future. Until such decision is taken it is recommended that Valente from NEA/CCDN act as contact. The nuclear data centres concerned should explore the most suitable ways of obtaining the information to be included in the list and of channelling it to the publishing centre on a regular basis.

In addition to the participants in this meeting, the list should be given a wide distribution particularly among users and producers of FPND.

To determine the distribution of the list outside the Panel the assistance of participants in this Panel should be solicited as well as of the Members and Liaison Officers of INDC, EANDC and other regional and national nuclear data committees. Until further notice all information in this respect should be sent to Valente.

(ii) It is recommended that an international newsletter on activities in the field of compilation and evaluation of FPND be developed as soon as possible. This newsletter should be published in regular intervals of 4-6 months. For each group or individual concerned it should list available manpower, names and addresses and contain a concise description of work finished, underway and planned and of recent publications and computerized data files with a brief indication of their format. Discrepancies in important FPND should be stated and also brought to the attention of the INDC Subcommittee on Discrepancies.

Noting that the international exchange of evaluated data is still restricted, the Panel proposes that as soon as possible, this recommendation be approved by INDC and brought to the attention of EANDC and other regional and national nuclear data committees.

The Newsletter should preferably be compiled and published by IAEA/NDS. It is however, left to the discretion of IAEA/NDS, NEA/CCDN and other nuclear data centres to decide upon this in the shortest possible delay after approval by INDC.

The newsletter should be distributed particularly to compilation and evaluation centres, and to groups and scientists working in the field of nuclear data, especially FPND.

(iii) It is recommended further that another separate international newsletter be developed covering measurement activities directly or indirectly related to FPND. Form and content of this newsletter should follow the model of the neutron capture γ-ray newsletter edited by G.A. Bartholomew and co-workers at Chalk River, Canada. For each experimental group it should contain a concise description of available facilities and manpower, of experimental work finished, underway and planned; it should list recent and forthcoming publications and give names and addresses of the scientists involved; it should also point to data discrepancies and specify standards used if suitable.

The newsletter should be published every 6 months and given a wide distribution particularly among measurers, but also compilers and evaluators of FPND. The aforementioned nuclear data committees and the participants in this panel should help to determine a suitable distribution. Also this newsletter should be published preferably by IAEA/NDS.

The approval of INDC for this newsletter should be sought as soon as possible and EANDC and other regional and national nuclear data committees should be informed of its decision. After INDC approval the nuclear data centres involved should decide who is to publish the newsletter, and its first issue should be published as soon as possible.

The Panel participants considered it suitable if, in addition to direct contacts, the Members and Liaison Officers of INDC would help to make sure that the contributions of their countries to the newsletter are provided regularly and on time to the publishing centre. For the time being IAEA/NDS will be the point of contact in all matters concerning this newsletter.

(iv) In order to improve the communication between users and producers of FPND it is recommended that the INDC at its next meeting in October 1974 discuss and approve the development of an international request list for FPND.

In order to assure that the list represent a realistic picture of the FPND requirements the Panel recommends that FPND requests should be justified by appropriate sensitivity studies and critically screened on the national scale before being submitted for international publication. These requests should be consistent with the Panel's findings, which emerged from discussion between users, measurers and evaluators of FPND.

In the compilation and publication of the list the existing WRENDA computer formats and intercentre cooperation should be used. The list should be updated and published by IAEA/NDS in annual intervals. The first issue should be published as soon as feasible after approval by INDC. The list should be given a wide distribution, particularly among nuclear physicists and measurers of FPND.

(v) In order to avoid a proliferation of computer formats the Panel recommends that the formats of FP yield and decay data as developed for the ENDF/B library and specified in Annex 1 with due regard of the deliberations presented in Annex 3 be adopted as the standard formats for the exchange of such data; for this purpose (A,Z) ordering should be used. For those institutions wishing to simplify that data for their own use some suggestions are given in Annex 2.

(vi) It is recommended that FP group cross sections (see Annex 2) be included in evaluated nuclear data files, provided that energy groups and spectrum used for averaging are specified in the same file.

(vii) Following the general discussion about information on experimental details needed by evaluators (chapter 1) the Panel recommends to improve the intercommunication between measurers and evaluators and to initiate a circular which would list information on experimental details, corrections applied, error analysis etc. evaluators require from FPND measurers. IAEA/NDS is to send a questionnaire to evaluators of FPND, asking them to state the information they want to obtain from

measurers. After having received the replies from evaluators, IAEA/NDS is to draft the circular and send it to evaluators for comment. The final, approved, version of the circular will be distributed to measurers. In addition, the circular will be included in one of the two newsletters recommended above.

It is realized that the inclusion of lengthy information on experimental details in publications may not be accepted by editors of scientific journals. Therefore it may be more appropriate to publish such information in laboratory reports, or send an information sheet directly to evaluators for their use.

(viii) The Panel noted that many observations and recommendations resulting from this meeting are of direct concern to the specialists group on nuclear data for applications to be convened by IAEA/NDS in Vienna from 29 April - 3 May 1974. This group will have the objective to coordinate on an international scale the compilation, evaluation, exchange and dissemination of nuclear level scheme and decay data of importance for applications in science and technology. The Panel recommends that all relevant observations and recommendations resulting from the present meeting including the data scope of FPND be given suitable consideration in the work of the specialists' group.

Annex 1

Definition of quantities in the ENDF format for FP yield and decay data

A detailed description of ENDF formats can be found in ENDF-102, Vol. I (last edition: BNL-50274, October 1970), which is generally available. Revisions have been made recently and are included in the data specifications given below. ENDF/B processing codes are described in ENDF-110 and the documentation of data is given in ENDF-201; both are also generally available.

Data type	Data specifications	Remarks
FP independent yields		FP yield data are listed for each individual fissile nuclide.
	- fission product identifier	- ZA, integer or real
	- isomeric state flag	- integer or real
	- yield	$\sum_i y_i = 2$
	- neutron energy or	- as a parameter (eV)
	- neutron spectrum specification	- recommended to give an evaluated point-wise spectrum
Radioactive decay data	General information:	
	- original nuclide identifier	- ZA, integer or real
including: $\alpha, \beta^-, \beta^+, \gamma$ isomeric trans. delayed neutrons	- isomeric state flag	- integer
	- half life of original nuclide	- in seconds
	- uncertainty of half life	- evaluated relative error, 1 standard deviation
	- number of average decay energies given	- integer
	- average decay energy for radiation x ($\bar{E}_x$)	- in eV; given in the order β, γ, α; delayed neutrons are presently included as β-decay
	- uncertainty of $\bar{E}_x$	- in eV

Data type	Data specifications	Remarks
	Decay mode information:	given for each mode of decay
	- total number of decay modes given	- integer
	- decay mode identifier	- real, included are γ, β^-, β^+, IT, α, delayed ns
	- isomeric stage flag for daughter nuclide	- integer or real
	- total decay energy (Q)	- in eV, Q-value available in corresponding decay process
	- uncertainty in Q	- in eV
	- decay branching (BR)	- fractional; given for radiation x
	- uncertainty of BR	
	Radiation spectra:	given as function of radiation energy for every decay mode
	- decay mode identifier	
	- number of spectra	- integer
	- radiation energy (E)	- in eV
	- uncertainty of E	- in eV
	- intensity of radiation (I)	- relative intensity, arbitrary units
	- uncertainty in I	- same units as I
	- internal conversion coefficient (ICC)	
	- uncertainty of ICC	
	- normalization factor (F)	- = ratio absolute intensity/relative intensity
	- uncertainty of F	- same unit as F
	- total number of energy points	

Annex 2

Specifications of FPND for applications

Data type	Application	Data specifications	Remarks
FP group cross section $\lceil(n,\gamma)$ or others$\rfloor$ (also for pseudo-FP)	burn-up long-range dynamics inventory calculation*	- Specification of group system, spectrum and method of averaging	} once for all FP
		- Nuclide identifier	- ZA, integer or real
		- cross section type indicator	- integer
		- group cross sections	
		- uncertainties of the group cross sections	- relative error
Independent yields of a FP nuclide	burn-up, long--range dynamics, decay heat inventory calculation*	- FP nuclide identifier	- ZA, integer or real
		- Identifier of fissionable nuclides having this FP	} ZA, integer or real
		- yields	
		- neutron energy or	- as a parameter (eV)
		- spectrum specification	- recommended to give an evaluated point-wise spectrum

- cont.

Annex 2
(cont.)

Data type	Application	Data specifications	Remarks
Decay constants	long-range dynamics inventory calculation*	- Daughter nuclide identifier - Identifier of nuclides decay of which leads to the above daughter nuclide - decay constants	- ZA, integer or real) ZA, integer or real - sec^{-1}
x-decay data (x=α,β,γ)	shielding design, safeguards, decay heat	- Nuclide identifier - Total decay energy (Q) - Uncertainty of Q - Average energy of radiation($\bar{E}x$) - uncertainty in $\bar{E}_x$ - Point-wise spectrum - energy of radiation x(E_x) - uncertainty of E_x - intensity of radiation x(I_x) - uncertainty of I_x - Normalization factor (F) - uncertainty of F	- ZA, integer or real - eV - relative error - eV - relative error - eV - relative error - relative - relative error - ratio absolute/relative intensity - relative error

Annex 2
(cont'd.)

Data type	Application	Data specifications	Remarks
Delayed neutron precursor data (Note: in ENDF delayed neutron precursor data are part of the decay data)	Reactor kinetics, safeguards fuel failure detection reactor operation	- Identifier of precursors - total decay constants - uncertainty of decay constant - cumulative yields of precursors - probability of neutron decay(Pn) - uncertainty of Pn - point-wise spectrum - delayed neutron energy (En) - uncertainty of En - incident neutron energy or - spectrum specification	- ZA, integer or real - sec.$^{-1}$ - relative error - fractional, absolute values - relative error - eV - relative error - as a parameter (eV) - recommended to give an evaluated point-wise spectrum

* As applied in shielding, reactor safety operation, fuel element endurance, etc.

Annex 3

Format Requirements for a library of evaluated FPND

A library of evaluated FPND has to contain:

i) FP yields;
ii) half-lives or the equivalent decay constant for the radioactive FP;
iii) beta and gamma ray intensities, energies and branching ratios for each FP;
iv) cross sections for each FP.

We shall discuss each item in detail, but first a few general comments are in order.

1. The data may be arranged either by FP nuclide, giving for each in turn yields, half-lives, decay data and cross-sections; or in blocks giving first all the yields then all the half-lives, then all decay data and finally all cross sections; or in some intermediate arrangement. Putting the data in blocks makes revision much easier as data from, say, a new evaluated fission yield library may be incorporated without much effort. Consequently, some blocking is recommended: certainly yields should be separate, and there are advantages in complete blocking.

2. It will be very useful to have some bibliographic information in the library, so that the sources of the data can be identified. Of course any user programme must then be able to read alphanumeric input.

3. One must always expect large amounts of data to have some punching errors, and often these may evade quite careful visual checking. In addition, faults and errors can occur on magnetic tapes during storage and use. To be able to detect such errors built-in checks are always desirable in large data libraries and suggestions for some will be made in the following sections.

Re (i): FP yields

Yields are needed for each fissile nuclide that is likely to be present in a reactor fuel, and either for several neutron spectra (e.g. thermal and fast power reactor) or at several specified neutron energies. In the latter case an interpolation scheme has to be assumed. For the input to an FP inventory programme, yields in reactor spectra are preferable but, for the exchange of data, specifying particular energies has the advantage of greater generality: but note that in this case the task of evaluation would be harder because most measurements of yields to date have been made in reactor spectra. Of course, a measurement in a reactor spectrum could be represented as a measurement at some suitable mean energy.

Either independent yields or chain yields and fractional independent yields may be stored. The latter alternative allows chain yields to be altered without having to re-punch fractional yields and permits extra checks to be made on the data: chain yields adding to 2 and fractional yields for each chain adding to unity. If independent yields are given they should sum to 2. Fractional independent yields for each chain of mass A may be expressed either explicitly for each Z or parametrically in terms of the most probable charge $Z_p(A)$ and the width $\sigma(A)$ of a Gaussian distribution: usually they are calculated from such a distribution.

Re (ii): Decay constants

Either half-lives or decay constants (in sec^{-1}) may be given. The former choice makes visual checking easier especially if each half-life is given in the most appropriate unit of time (sec, min, hour, day or year) with the rule that the unit be chosen so that the numerical value of the half-life is as small as possible but not less than 1. The unit could either be specified by a numerical code or by its initial letter: the latter allows easier checking but requires slightly more complicated programming. Although the ENDF format at present requires the half-life to be given in seconds, a blank field is available in which a unit indicator could be specified.

Re (iii): Decay schemes

The essential data are (i) average beta energies, (ii) gamma ray energies and intensities and (iii) branching ratios to alternative product nuclides or isomeric states. No existing programme calculates a beta energy spectrum for gross fission products and it is most unlikely that this will ever be needed, so that average beta energies are adequate. The French library gives beta end - point energies and probabilities, so the data have to be processed by an auxiliary programme to obtain the average energy. One small disadvantage of this refinement is that presumably all the transitions are assumed to be allowed, as no information is stored about the classification of each beta decay.

For gamma-rays a spectrum is needed, for use in shielding calculations. One can group gamma energies in "bins", or give individual energies: this latter more general convention seems preferable as the specification of the "bins" may need changing as photon transport codes become more powerful. Conversion electron and X-rays should be given in a special "bin" or line.

Intensities for individual transitions may be given either absolutely or relatively.

It is worth pointing out that none of the libraries available contains all the data available in a full decay scheme. This would involve giving, for each level of the product nucleus, its spin and parity and energy above the ground state and the probability of decay to it from the parent nucleus and of decay from it to each lower level. However, such extra-complexity would not give any extra useful information to FP inventory programmes.

It is necessary to have a rule for defining "isomeric state". The compilers of ENDF assume that no level with a half-life less than 0.1 sec. need be considered separately as an isomeric state: this limit appears quite adequate.

Re (iv): Cross sections

Inventory programmes need (n,γ) cross sections for thermal, resonance and fast groups. On the other hand, ENDF/B, being part of a larger library, gives only point cross-sections for all relevant reactions. However, few-group cross-sections will depend on reactor spectrum, and so it will be desirable to be able to change them easily. Consequently, if ENDF is adopted as standard, it is suggested the the FP library contain, as an additional separate block, well defined few-group capture cross-sections. The details of a proposed format can be discussed later.

Uncertainties

ENDF allocates space for uncertainties in half-lives, decay data and branching ratios. This is an interesting and potentially useful development, which is very much appreciated by FPND users. The inclusion of yield data uncertainties should also be developed for ENDF. For problems such as the calculation of the FP decay heat and its error, the Panel considers the knowledge of FPND uncertainties as indispensable.

Conclusions

ENDF fits the requirements better than the other formats and as it also has advantages in the international exchange of data, its use as standard is recommended. It may be possible to make a few changes in it to make it match up even better to the requirements.

Consideration of possible checking programmes should begin as soon as possible. A short list of checks that could be made follows:

a. Yields should add to 2.
b. $\bar{\nu}$ should be calculated, from the yields, for comparison with recommended values: in addition, the mean atomic number should equal $Z/2$, where Z is the atomic number of the fissile nucleus.
c. Partial decay energies should sum to the total available.
d. Branching ratios should add to unity.

3. FP INVENTORY AND DECAY HEAT

3.1. General subdivision of data requirements

3.1.1. Observations and conclusions

(i) FP inventory is the amount of individual and collective fission products present in a fuel element or part hereof, or in the reactor core, at any time during or after irradiation. FP inventories are required in nearly all areas connected with the nuclear fuel cycle, as covered by RP's 2-7 at this meeting. FPND required for the determination of inventory are:

- FP yields
- neutron absorption and - capture cross-sections
- half lives
- decay branching ratios

These data are referred to as "inventory data" in the following.

(ii) In some cases only the knowledge of certain bulk properties of mixed FP is required and the inventory of individual FP can be replaced by that of groups of FP, e.g.:

- total FP absorption: individual FP are replaced by pseudeo FP;
- delayed neutrons: the total delayed neutron yield or delayed neutron groups are sufficient in most cases;
- total FP energy release: the concept of pseudo FP needs further investigation

(iii) In some fields of application further decay data in addition to inventory data are required for the calculation of certain properties of a mixed FP source

- For the calculation of the total energy released by FP after reactor shutdown, either the average β- and γ-energy emitted per decay, or the effective decay energy of (important) individual FP is required.
- The knowledge of penetrating radiation emitted by FP is needed in shielding.

(iv) Finally, decay properties only are required for the measurement of individual FP.

- Properties of the characteristic radiations emitted by FP have to be known for their identification and for the determination of their content in a sample.
- A number of decay characteristics of radioactive species are required in all fields where the interaction of radiation with matter is important, such as life sciences, industrial and agricultural applications.

3.2. Needs for inventory data and bulk properties of FP

3.2.1. Observations and Conclusions

The Panel noted that in general accuracy requirements for bulk properties of FP, such as total FP absorption or energy release, or for the FP inventory have been well assessed in the review papers to this Panel. However, a great deal of more work is needed to assign accuracy requirements to individual FPND consistent with those for bulk properties. In particular, accuracies already achieved for available FPND should be taken into account.

(i) Environmental and exposure studies require a rather complete knowledge of FP inventories, starting from FP half lives as short as <1 s in cases of accidents and nuclear explosions. No studies have yet been performed to evaluate the accuracy to which the FP inventory is required. Since a vast number of FPND is involved in this field and a lot of information on these FPND is already available, it is the feeling of the Panel that

- theoretical studies should be performed as to which FP constitute an important hazard and to what accuracy the knowledge of their inventory is required;
- the needs for individual FPND should be evaluated in the light of the accuracy presently achieved;
- with respect to the importance of FP with yet unmeasured properties, limitations on the ranges of yields, half lives and Q values should be given on the basis of theoretical considerations.

(ii) The Panel endorsed the conclusions of J.G. Tyror (RP 3) that is is necessary to

- achieve a target accuracy of 2% in the prediction of the fuel reactivity life times due to fission products alone for thermal reactors;
- evaluate the effect of fission products in a typical fast breeder reactor to within 0.5% of reactivity, i.e. to within 10% accuracy in FP captures;
- evaluate the change of reactivity held by fission products due to the Na-void effect to within 0.2% of reactivity in fast reactors, i.e. to evaluate the change of FP captures to within 30%.

Tyror pointed out that his target accuracies for individual FPND, listed in Tables IV and VIII of RP 3, were obtained as one possible and economically justifyable solution among others for achieving the accuracy requirements for total FP capture quoted above. The Panel accepted these target accuracies, as reproduced in Appendices A1-A4, but recommends the study of other solutions based on the accuracies which may be obtained both from experiments and theoretical calculations.

(iii) For FP release and contamination of reactor components, the Panel, after some discussion, adopted C. Devillers' interpretation (RP 4) of R.H. Flowers' proposal of FPND requirements. These accuracy targets for the general case without using any knowledge of the presently available accuracy of the required FPND are (1 standard deviation):

inventory to $\pm$ 40%, comprising:

fission yields to $\pm$ 20%

half lives to $\pm$ 5%

neutron capture cross sections sufficiently accurate to allow a calculation of the term $(\lambda + \sigma\phi)$ to $\pm$ 5%.

The last requirement should hold for fluxes of about 10^{14} and 5×10^{15} neutrons$\cdot$cm$^{-2}\cdot$sec^{-1} for thermal and fast systems, respectively. The Panel concluded that for stable FP 20% accuracy on the capture cross-section would be sufficient.

Individual figures again depend on the presently available accuracy of FPND. As can be seen in Appendix A3, the uncertainties of half lives are much less than 5% in most cases and can in fact be neglected compared to the 40% uncertainty required for the inventory. Thus the tolerable error of fission yields can be raised to about 40% provided that neutron capture is insignificant. The significance of capture cross sections, on the other hand, needs further investigation in some cases.

(iv) For failed fuel detection the Panel adopted the accuracy requirements presented in RP 4 for the general case (see Appendix A3). Again, half lives are generally known more accurately than required and accuracy targets should be reconsidered as said under (iii) above.

(v) Fuel element design was not foreseen to be included in any review paper, but data requirements were discussed briefly during the meeting. The technical background and FPND requirements are taken from a contribution to RP 4 by R.H. Flowers with additions from the Panel discussions:

In fuel element design, chemical and mechanical interactions arising from FP present in a burnt fuel element have to be considered, specifically:

- calculation of noble gas pressure within the fuel,
- calculation of oxygen potential changes due to replacement of U or Pu by FP,
- calculation of volume changes in fuel.

General ND requirements for FP with half lives > 1 day are (1 standard deviation):

- FP inventory: to $\pm$ 20% for FP with cumulative yields $\geq$ 1%,
  to $\pm$ 50% for FP with cumulative yields between 0.1 and 1%,
  within a factor 5 for FP with cumulative yields < 0.1%,
  comprising:
- FP cumulative yields from
  thermal fission of ^{233}U, ^{235}U, ^{239}Pu and ^{241}Pu and from
  fast fission of ^{232}Th, ^{233}U, ^{235}U, ^{238}U, ^{239}Pu, ^{240}Pu and ^{241}Pu
  to $\pm$ 15% for FP with cumulative yields $\geq$ 1%,
  to $\pm$ 50% for FP with cumulative yields between 0.1 and 1%,
  within a factor 5 for FP with cumulative yields < 0.1%;
- branching ratios sufficiently accurate to allow calculation of the cumulative yield of daughter products within the limits given above;
- half-lives and capture cross-sections: the term $(\lambda + \sigma\phi)$ should be known accurately enough to calculate the inventory within the limits given above. Neutron fluxes to be considered and arguments for splitting up accuracy requirements between T1/2 and σ are the same as those stated under (iii) above for FP release and contamination of reactor components. Accuracies of capture cross-sections of stable FP should meet requirements for inventory after burn-ups up to 90% FIMA.

Adequate calculation of the gas pressure within a fuel element due to noble gas FP requires more accurate data:

$\pm$ 10% are required for the inventory of stable rare gases, comprising
$\pm$ 5-10% for cumulative fission yields (= chain yields), and

± 5% for neutron capture cross-sections (if relevant)

The fission yield requirements listed are for thermal fission of ^{233}U, ^{235}U and ^{239}Pu and fast fission of ^{235}U and ^{239}Pu. Accuracy requirements are lower for other fissionable isotopes listed above.

FP half-lives exceeding 1 day are generally known accurately enough to be negligible against the other uncertainty limits given above. Thus the accuracy requirements for yields $\geq$1% can be raised to ± 20% if neutron capture is insignificant. Furthermore, for FP with half lives > 1 day total chain yields can be used together with branching ratios within the requested accuracy limits.

(vi) The Panel agrees that for fuel handling generally the total heat released by FP should be known to ± 5% or better from about 3 months onwards. In future, this accuracy should be reached already from about 1 month cooling time onwards as needed for Pu recycling of fast-breeder reactor fuel. Needs for energy released by FP are summarized in Appendix A5.

The Panel accepted for its present survey the FPND requirements presented in RP 7 which are based on more general considerations. It recommends, however, to re-evaluate accuracy requirements for individual FPND with the aid of Devillers' decay heat studies, using available FPND. These studies became available only after the Panel meeting and are included as appendix to RP 4 and presented graphically in Appendix A5.

Data requirements for FP constituting a potential hazard in fuel handling are generally agreed by the Panel.

(vii) Nuclear fuel burnup can be determined directly using long-lived stable FP as burnup monitors (destructive and non-destructive analysis), or indirectly (correlation studies) with the aid of ratios of the number of FP atoms (destructive analysis) or of FP activities (non-destructive analysis). Details are given in RP 5 and 6. Accuracy requirements for burnup determination presented to the Panel ranged from about 2% (USA, EURATOM) to 5% (France, UK, USSR). After some discussion the Panel agreed on the FPND requirements presented in the chapters 4,5 and 7, which imply that

- burnup can be determined to about 3% by methods presently employed for destructive analysis;
- requested FPND accuracies are just adequate for non-destructive analysis; the implied uncertainties do not constitute a major source of error.

The Panel noted that generally ^{148}Nd, determined by destructive methods, is the most suitable and widely used burnup monitor. Other burnup monitors are required for practical cases where ^{148}Nd is less suitable. Burnup determination by non-destructive methods is gaining importance, since it is less expensive.

The short-lived (compared to fuel irradiation time) FP ^{95}Zr-Nb, ^{103}Ru, ^{140}Ba-La and ^{141}Ce are used for the determination of burnup after short-term irradiation and of fission rates within fuel elements prior to discharge (rating measurements). Generally, such measurements are performed employing non-destructive analysis and, compared to long-lived stable FP, uncertainties are increased by the complexity of irradiation histories. The Panel agreed that for these determinations an accuracy of 5% should be achieved; this implies the FPND requirements presented in Appendices A1, A3 and A4.

a) FPND requirements for burnup monitors are well defined. For the determination of burnup primarily fission yields of burnup monitors are required. For non-destructive analysis, half-lives and absolute γ-ray intensities are needed in addition. In order to derive the burnup from the measured inventory of a monitor FP, corrections for build up and burn-out of this FP due to neutron capture may have to be calculated. Uncertainties due to these corrections should be $\lesssim 1\%$ of the burnup. The requirements for the cases discussed below are given in chapter 7 and Appendices A1 and A4.

- The isotope ^{142}Nd is not formed in fission. Therefore the amount of ^{142}Nd found in a mass-spectrogramme is used to correct for contamination by naturally occurring Nd in a destructive measurement of FP Nd. Prior to this correction the ^{142}Nd formed by neutron capture in ^{141}Pr has to be subtracted. Presently available thermal fission yields and the thermal capture cross-section of ^{141}Pr are adequate. No requirements have yet been specified for fast burnup.
- At high burnups from irradiations in high thermal neutron fluxes a non-negligible amount of ^{148}Nd is formed via neutron capture in ^{147}Nd. The Panel noted that the capture cross-section of ^{147}Nd was still unmeasured, while available data for fission yields and the half-life were found to be sufficiently accurate for corrections.
- Further corrections may have to be applied for buildup and burnout of burnup monitors. The accuracies requested for fission yields of burnup monitors are sufficient for these corrections and needs for neutron capture cross-sections are discussed in chapter 7.

b) The situation is different for isotope correlation studies where the method is based on a comparison of measured and calculated ratios of FP atoms or activities. The Panel noted that the only isotope ratio used routinely for burnup determination is that of the neutron capture product ^{134}Cs to ^{137}Cs. FPND requirements expressed by the Panel are based on RP 5 and 6, and results of a sensitivity study by Foggi and Ley (ref [46] quoted in RP 6).

(viii) Safeguards: FPND requirements for post-irradiation fuel analysis are mainly covered by burnup requirements, but FP isotope ratios (correlations) are used more extensively in safeguards in order to derive additional information such as cooling time, ^{239}Pu buildup etc. (see RP 6). However, the usefulness of these methods for routine investigations in safeguards remains to be proven. More work has to be done to assess quantitatively the accuracies that can be achieved with the procedures for analysis of spent fuel outlined in RP 6 and their impact on FPND requirements.

The FPND that have to be known and their present adequacy for safeguards are summarized below as observed by the Panel:

- Most of the FP used for ratios are also burnup monitors and the FPND accuracies requested for burnup are adequate for safeguards.
- ^{154}Eu, formed by neutron capture in ^{153}Eu, is potentially useful for the determination of burnup, fluence and ^{239}Pu fissions. FPND accuracies for mass chains 153 and 154 requested in RP 5 for burnup determination correspond to a tolerable uncertainty of $\sim 3\%$ in the ^{154}Eu inventory, and hence also in the ^{153}Eu invertory. This implies that the amount of ^{153}Sm removed by neutron capture has to be known to 3% accuracy. Since the half life of ^{153}Sm is known to $< 1\%$, the term $\sigma\phi$ should be known to $\pm$ 3% of $(\lambda + \sigma\phi)$. Requirements for the unknown capture cross-section of ^{153}Sm are discussed in chapter 7, other requirements taken from RP 5 are given in Appendices A1, A3 and A4.

  However, the Panel noted that also mass chains 149, 151 and 152 contribute significantly to the ^{154}Eu inventory at high burnup levels through multiple neutron capture, as shown by Eder and Lammer (reference [3] quoted in RP 6). Capture cross sections and fission yields are required in addition for these mass chains, but the accuracies needed are not yet known.
- Isotope correlations using ratios of Kr, Xe and Nd fission products have so far been studied purely empirically (see RP 5 and 6). Much more work is required, including calculations and sensitivity

studies, in order to understand the processes involved, develop the method to practical applicability and specify FPND requirements (see RP 6).

For fresh fuel assay a complete library of FPND including FP with half-lives down to 1 sec are of greatest importance.

3.2.2 Recommendations:

(i) The Panel recommends that investigations be performed in all user areas, where this has not yet been done, aiming at a detailed specification of needs for individual FPND and their accuracies. Such investigations should yield the following information:

- identification of FP important in the area concerned; criteria for significance of FP with respect to yet unknown properties;
- accuracy requirements for bulk properties of FP or for FP inventory data;
- needs for individual FPND backed up by sensitivity studies; presently available accuracies of FPND should be used, as observed by the Panel, or taken from more recent evaluations.

These types of investigations should not only be performed to back up a WRENDA-request (see Chapter 2) but form the basis for review papers and discussions of the proposed follow-up meeting on FPND. The Panel recommends, that studies be initiated as soon as possible.

(ii) When assessing the needs for individual FPND, users should also take into account:

- whether higher accuracies for some FPND are requested in other application fields with high priority, and
- which sub-division of FPND needs might be least expensive to be satisfied.

The appendices on comparison of FPND status and requirements may help in these decisions.

3.3. FP decay heat

3.3.1. Observations and conclusions

(i) Total energy released after reactor shutdown ("afterheat"): In three different fields of application a knowledge of the decay heat generated within a fuel after reactor shutdown is required:

- residual power as a function of time after emergency shutdown of a reactor (in case of an accident), starting from zero up to a few days cooling time (RP 4);
- fuel handling and intermediate storage at the reactor site, starting from 8 hours up to a few months cooling time (RP 4);
- fuel transport, reprocessing and waste disposal, starting from about 1 month cooling time (RP 7).

The latter is discussed in the previous section. For the other fields which are covered by RP 4, C. Devillers gives a survey of different user needs. The Panel affirms the urgent need to improve the accuracy of the afterheat function in order to save unnecessary deratings of reactors. The accuracy requirements for the total heat released as a function of time has been assessed by the Panel in all 3 areas and is presented in Table A5-Ia of Appendix A5.

(ii) FP contribution to total afterheat: The Panel endorses the conclusions of C. Devillers (RP 4):

FP energy release has to be known primarily from thermal fission of ^{233}U, ^{235}U and ^{239}Pu and from fast fission of ^{235}U and ^{239}Pu (see Table A5-Ib in Appendix A5). Of secondary interest are contributions from ^{241}Pu thermal fission (recycled Pu) and fast fission (10-20% contribution to total fissions) and ^{238}U fast fission (about 1/3 of the accuracy required for primary fissile nuclides).

However, it has been observed (RP 4) that the calculated afterheat changes by $\lesssim 1\%$ only if one replaces ^{239}Pu thermal fission yields by fast fission yields. Therefore the knowledge of fast fission yields appears to be not of primary importance.

The contribution of FP to the total decay heat is $\lesssim 40\%$ up to 1 sec cooling time, $\lesssim 50\%$ at 10 sec and $\sim 90\%$ from 100 sec onwards. This explains the accuracy requirements for the total energy released by FP as summarized in Appendix A5 (Table A5-Ib).

(iii) Calculations and measurements of FP decay heat: M. Lott has reviewed (RP 15) existing calculations and measurements of energy released by FP. The conclusions of Lott, Devillers (RP 4) and the Panel are summarized below.

Comparison of calculations:

Essentially two methods are used to calculate the total FP energy release. In one approach the total decay heat is represented by an analytical formula which could be derived only from highly discrepant measurements (K. Shure, see RP 15 for details). In the second and most

widely applied approach the decay heat is calculated from inventories of individual FP and their energy release per decay. Only this latter method needs FPND and is referred to as "calculations" in the following discussion.

At short cooling times ($\lesssim$ 10 sec) the calculations compared in RP 4 and 15 tend to under-estimate the heat release, particularly after short irradiations. It can be concluded that this is due to very short-lived ($\lesssim$ 1 sec) FP with unknown ND which are, however, less significant at the end of irradiations in reactor operations. Measurements would be required in order to fill this gap, but rather large uncertainties of individual FPND could be tolerated in order to reach the required accuracies of 25% at 1 sec and 20% at 10 sec cooling time (see Table A5-Ib, Appendix A5) for the total FP heating, as due to the very large number of contributing FP a partial cancelling of the uncertainties can be expected. On the other hand, the number of "unknown" FP increases with decreasing cooling time. The use of an improved analytical expression, derived from new consistent measurements of FP decay heat, might be more suitable below $\sim$1 sec cooling time. Further investigations are therefore necessary to show whether and which new measurements of FPND are required and/or whether the unknown FP could be replaced by a lumped short-lived FP for certain cooling times.

At cooling times of $\sim 10^2 - 10^3$ sec individual FP are more significant and the calculations partly disagree. Available FPND are not sufficient and improvements are required.

FPND required for cooling times above 10^3 sec are generally well known from measurements. Calculations agree well between 10^3 and 10^7 sec . Above 10^7 sec, where only few FP are important, discrepancies among calculations could be resolved by a comparison and critical re-evaluation of the input data used.

Apart from some general statements about accuracy requirements for FP decay heat the Panel noted only deficiencies in particular FPND as included in the data libraries compared in RP 15, but no studies on their significance were available. Therefore the Panel recommended an immediate action on the French group (Devillers, Lott) who surveyed the subject for the meeting, to prepare a list of important FP dominant at each cooling time of interest for inclusion in the Panel proceedings (see (iv) below).

Comparison with measurements:

Discrepancies exceeding individual experimental errors between the re-

sults of different FP decay heat measurements are observed, which exceed by far those between calculations. The Panel noted that these discrepancies cannot be resolved as the different experimental conditions do not allow a direct comparison. Nor can the reliability of the calculations be checked in a meaningful way against discrepant measurements.

Benchmarks:

Existing discrepancies among experimental results and possible systematic errors can only be resolved if new measurements, employing all available methods, are performed under identical irradiation conditions; independent results could be obtained if such benchmark experiments would be performed at different laboratories. After collection and proper evaluation, these results could serve to derive an improved analytical expression for the heating function.

In order to allow a comparison between different calculations and between calculation and experiment, benchmark calculations should be simultaneously performed at pertinent laboratories for the irradiation conditions of the experiments and for a wide range of cooling times. Uncertainties of FPND should be incorporated in the FPND libraries used in these calculations and the uncertainty of the total FP decay heat calculated in each individual case.

The analysis of the results of experiments and calculations should yield the following information:

- It should be possible to check the reliability of the calculations and derive an overall uncertainty of the afterheat function.
- The analysis of agreements and disagreements between measurements and calculations for different cooling- and irradiation times should help to check uncertainties and to identify significant deficiencies of input data used in the calculations.
- The comparison between all decay heat results at very short cooling times should help to determine whether or not ND of new short-lived FP, and which ones, have to be measured and included in FPND libraries.
- Finally, these benchmarks should enable to establish a detailed list of FPND requirements for "afterheat" for all cooling times.

(iv) Upon the recommendation of the Panel, C. Devillers has provided a list of FP contributing $\leq 1\%$ to the total FP energy release at cooling times of 10^0, 10^1, 10^2 ... 10^9 sec for 5 different practical cases (including ^{233}U, ^{235}U, ^{239}Pu thermal and ^{239}Pu fast fission). The results are presented in detail as appendix to RP 4, and all 5 cases are combined in

a graphical representation shown in Appendix A5, Table A5-II. The calculations confirm the Panel's conclusions in several points:

- Up to ~100 sec there is a large fraction of FP contributing less than 1% to the total FP decay heat.
- No FP contributes more than 4% to the total FP decay heat up to ~100 sec.
- Only few FP are significant above about 10^6 - 10^7 sec.

(v) The results of Devillers' and Vossebrecker's* investigations can be combined with the Panel's findings to the following conclusions:

- The present knowledge of FP chain yield data appears to be adequate for decay heat calculations. It should be sufficient to check the influence of the difference between thermal and fast fission yields for ^{235}U and ^{241}Pu.
- Fractional cumulative yields can probably be derived adequately from empirical charge distributions, as discussed in chapter 4. However, this point should be further investigated by sensitivity calculations.
- Requirements for decay data can be given qualitatively at least for the dominant FP identified by Devillers.
- The present knowledge of neutron capture cross-sections is sufficient for decay heat calculations (see RP 4).
- Rather large statistical uncertainties of individual FPND can be tolerated below ~100 sec cooling time.
- Detailed specifications of FPND requirements have to await the completion of benchmark experiments and calculations. Therefore no detailed comparison of FPND status and user requirements for after-heat has been performed by the Panel.

3.3.2. Recommendations

(i) Although the FP decay heat values obtained from different calculations agree well for cooling time between 10^3 and 10^7 seconds, it is not possible to draw any definite conclusions on the uncertainty of the after-heat function from this agreement, as the sources of data are often the same. Therefore the Panel recommends that error bars should be included in libraries of FPND. Errors should also be estimated for theo-

* Stimulated by this Panel, H. Vossebrecker (INTERATOM GMBH, Bensberg, Köln, FRG) has recently calculated uncertainties of the FP decay heat at various cooling times from FPND uncertainties. The results were communicated to the Scientific Secretaries of the Panel and are summarized here since they are of interest in the context of the Panel's findings:

- The calculations confirm that considerable FPND uncertainties can be tolerated at short cooling times.
- Uncertainties of fission yields do not contribute significantly to the overall uncertainty of the decay heat.

retically derived ND values and new calculations of afterheat functions and their uncertainties be performed.

(ii) Internationally coordinated benchmark experiments on FP energy release should be performed at different laboratories. These measurements should be performed at given cooling times following irradiations at constant sample power. The irradiation conditions should be as identical as possible. The methods involved are:

- calorimetry should be used wherever possible to serve as reference method;
- other methods should be applied where calorimetry cannot be used, i.e. above 60-100 seconds;
- above 100 seconds all methods should be used at given reference points to allow an intercomparison.

The precision of these measurements should be as high as possible.

Parallel to these benchmark experiments and for the same measurement conditions, coordinated afterheat calculations, including estimates of uncertainties, should be performed at different laboratories. This would allow a better and direct comparison between different calculations and experimental results and would give a first global test of the reliability of FPND libraries.

The results of all benchmark experiments and calculations should be collected, analyzed, evaluated and published together with conclusions in a final report.

The Panel considers it important to have results of the benchmark experiments available as soon as possible. Therefore the Panel recommends to start with first priority measurements on ^{235}U thermal fission, which is simpler to perform, and only on a restricted range of cooling times. ^{235}U could then serve as a standard for further measurements. Benchmark experiments on ^{239}Pu thermal fission are proposed with second priority.

The Panel recommends that IAEA/NDS acts as the point of contacts and organizes the benchmark experiments. Lott will work out the guidelines with respect to experimental conditions and methods to be used and send them to IAEA/NDS for distribution. He will also supply IAEA/NDS with a list of laboratories and/or scientists that could participate in benchmark experiments. IAEA/NDS will work out a time schedule together with Lott and takes the responsibility for contacting other laboratories and coordinating the experiments. Lott will collect the results of the benchmark experiments and organize the evaluation and conclusion of the intercomparison. IAEA/NDS will assist him in the publication and distribution of the final report.

(iii) No means has yet been established to communicate information on finished or on-going studies related to afterheat. In order to fill this gap, and particularly for the benefit of the reviewer of this subject for the following meeting, the Panel recommends that all such information be communicated to IAEA/NDS for collection and transmission.

(iv) Although a detailed formulation of user requirements will have to await the completion of the benchmark experiments, some actions should follow Devillers' study of FP important for afterheat:

- The tolerable overall uncertainty of the remaining FP can be derived with the aid of the presently available status of the listed FP.
- More detailed FPND requirements can be worked out for cooling times exceeding a few hours ($\sim 10^4$ sec)
- Evaluators could concentrate on updating the status of nuclear data of important FP and communicate poorly known FPND to measurers via the newsletter proposed in Chapter 2.

4. FP YIELD DATA

4.1. Chain yields

This section deals with fission products whose cumulative yields are essentially identical to the total chain yields.

4.1.1. Observations and conclusions: user requirements

(i) The Panel agreed that burnup measurers would be satisfied at present with an accuracy of 2% in the yields from the major fissile isotopes with a long-term goal of 1%. The yield accuracies required for short lived FP used in non-destructive fuel analysis were considered to be not as stringent in view of the lower accuracy of the methods concerned.

The Panel noted that at present ^{148}Nd is the most suitable burnup monitor that is commonly used for different fast reactor fuel types. However, it has been noted in RP 5 that the variation of its fission yield with the median energy of fast reactor spectra (expressed as spectral index in RP 5) may exceed the requested accuracy, particularly the long term aim of 1%. Therefore the variation of the ^{148}Nd fission yield with incident neutron energy has to be known

to the accuracies specified in Appendix A1 in order to achieve the required burnup accuracies. As any determination of ^{148}Nd either for burnup or in a fission yield measurement involves a determination of all FP Nd, the Panel recommends to extend measurements of the energy dependence of yields to all Nd isotopes. This should enable users to select the most suitable burnup monitors.

(ii) Thermal yield requirements for fuel design are met by the available data (see Appendix A1).

In the case of fast reactors the requirements are separated into those for the calculation of the gas pressure within a fuel element, and those for the investigation of chemical interactions of FP with the fuel and cladding material. The former requires yields of stable rare gas FP only.

For investigations of the chemical state of fast reactor fuel, the knowledge of complete mass yield curves is required. This is primarily of importance for fuel design and development. However, such investigations are also of interest for burnup, as information on diffusion, migration and volatility of FP influence the selection of burnup monitors for different types of fast reactor fuel. Requirements for fuel design contributed by R.H. Flowers are presented in section 3.2 (item (v)). Independently, this topic was discussed by the subgroup on chain yields. Requirements expressed by Flowers and the subgroup are presented separately in Appendix A1.

(iii) All further requirements for FP yields as presented in review papers and agreed by the Panel are summarized in Appendix A1. They are, however, subject to the limitations discussed in chapter 3 (inventory data). In particular, the Panel noted that certain yields can be estimated within an uncertainty margin of 10-30% by interpolation or calculational methods, which would satisfy some user requirements. Since accuracy objectives for bulk properties (e.g. total FP captures (RP 3) and FP decay heat (RP 7)) can be achieved by different ways of allocating requirements for individual FPND, the study of other solutions is recommended on the basis of available yield uncertainties and the capability of estimating unmeasured yields.

(iv) Stable and long lived FP contribute to the production of photoneutrons via capture- and decay γ-rays. Although this topic was not discussed in detail by the Panel, it should be noted that the knowledge of FP chain yields is required, and detailed needs should be investigated.

4.1.2 Observations and conclusions: status of FP chain yields

(i) Thermal fission yields

From the information presented in RP 11a and the data given in Appendix A1 it appears that the majority of the requirements for thermal fission yields has been met with the exception of a few cases where discrepancies among experimental data and larger uncertainties still exist, as shown in more detail in Appendix A1. However, the Panel did not consider it economically feasible to initiate additional extensive measurement programmes for the determination of thermal yields. Evaluators should rather try to resolve discrepancies by careful examination and selection of existing experimental data and recommend some limited less expensive measurements if deemed necessary.

(ii) Fast fission yields

The Panel noted that "fast" yields of certain FP important for users depend on incident neutron energy in the range of interest for fast reactor applications (see RP 5 and 11b). Therefore, user requirements have to be understood, at least in principle, as being expressed for yield data as a function of fast reactor spectrum. However, the Panel concluded that a term "fast yields" should be maintained and associated with a set of yield data for the present survey (Appendix A1) as well as for future considerations for several reasons:

- In the past, reactor neutron spectra used in fission yield measurements were generally not defined. Consequently, fast yields obtained in current evaluations, from which the data status is drawn, are not well defined either. Therefore the status of fast yields reviewed by the Panel has to be used in this sense and is explained in detail in Appendix A1.
- For most fast reactor applications the change of yields with neutron energy is well within the requested accuracy limits. Even for the most stringent requirements, the variation of yields in the mass peaks is expected to be tolerable.
- Future large fast reactor power stations will have rather similar neutron spectra.
- Therefore it is desirable that also in the future "fast yields" be evaluated, although for defined neutron spectra, which may be associated with some kind of information on the energy dependence of yields. In the transition period, fast yields have to be used as they are defined by evaluators.

Yield measurements will continue to be performed in fast reactor spectra and will most likely be considered as one separate group of data by evaluators. The classification "fast yields" will be used for this type of measurements, but information on the neutron spectrum in which yields were measured should be included in publications. The way in which fast yields will be presented by evaluators depends on their individual approach to the problem, but it would be desirable to relate the definition of "fast yield" with neutron spectra of commercial fast breeder reactors.

A number of requirements for fast yield data have not been attained; however, extensive measurement programmes are in progress in various Member States. These programmes, the expected accuracies to be attained and the projected completion dates are outlined below:

Euratom (Koch)

Absolute yields of Cs, Ba, Ce, Nd, Sm and Eu isotopes, and relative yields of Kr and Xe isotopes are being measured in fission of ^{232}Th, ^{233}U, ^{235}U, ^{236}U, ^{238}U, ^{237}Np, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu, ^{241}Am and ^{243}Am irradiated in the French reactor Rapsodie. The work on ^{235}U, ^{238}U, ^{237}Np, ^{239}Pu, ^{240}Pu and ^{241}Pu will be completed in 1974, the remainder before 1977.

France (Bouchard)

Measurements are being made of the yields of all Nd isotopes from the fission of ^{235}U, ^{238}U, ^{239}Pu, ^{240}Pu and ^{241}Pu irradiated in the fast reactor Rapsodie and Phenix. The measurements in Rapsodie are almost finished, while those in Phenix should be completed in 1975. Nd yields in fission of ^{238}Pu, ^{242}Pu and ^{241}Am are also being measured in Phenix.

India (Ganguly)

Radiochemical measurements of fast yields of nuclides in the wings of the mass yield curves from the fission of ^{233}U, ^{235}U and ^{239}Pu are being made. Expected completion date 1976.

Switzerland (von Gunten)

Radiochemical measurements by γ-spectrometry of yields of nuclides from mass 87-105 and 129-151 are being made for fission of ^{235}U and ^{239}Pu in the Proteus reactor which has a neutron spectrum similar to a Helium cooled fast reactor. The experiments have been completed and the accuracy achieved is 2-5%. The final results have been submitted to Nucl.Sci.Eng.

U.K. (Sinclair, Crouch)

Mass spectrometric measurements of fission yields of all Nd isotopes and of some long lived isotopes from fission of ^{235}U, ^{239}Pu, ^{240}Pu and ^{241}Pu, irradiated in the DFR reactor, are being made by Sinclair.

The work should be completed by 1976.

Similar measurements are being made by Crouch for fission of ^{235}U, ^{238}U, ^{239}Pu, ^{240}Pu and ^{241}Pu irradiated in the DFR reactor; the first three of these should be completed by 1975, the others will start in 1974. He is also carrying out a similar series of irradiations in PFR, starting in 1974.

U.S.A. (Maeck)

Mass spectrometric measurements of yields of the entire mass yield curve are being carried out on samples which have already been irradiated in EBR II. Measurements have been completed for ^{235}U and ^{238}U, but have still to be done for ^{233}U, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu, ^{241}Am, ^{243}Am and ^{237}Np. Samples of ^{233}U, ^{235}U, ^{239}Pu and ^{241}Pu were irradiated in two different EBR II spectra so as to examine the effect of neutron energy.

USSR

Measurements are in progress on the reactors BOR-60 and BR-5 in connection with an investigation programme for fast reactor physics and burn-up. Yield measurements using mono-energetic fast neutrons are also being carried out on a number of fissioning nuclides.

It is possible that these measurements will be sufficient to satisfy user requirements after they will have been reported and evaluated, but the Panel feels unable to express this possibility in a quantitative way.

(iii) Effect of neutron energy on fission yields

Systematic studies of the effect of neutron energy on yields should be performed at least in the mass ranges 103-125 and 140-155. For other chain yields it should be sufficient at present to check the extent of the energy dependence. The ultimate goal would be the derivation of an expression that associates yields with neutron energies. The important function of evaluators is to correlate measured yields with neutron energy, identify significant changes and find the most suit-

able way to describe fast reactor spectra in terms of an energy dependent parameter.

Several ways are used to characterize fast reactor spectra:

- The spectrum can be given point wise or in energy groups.
- An average or median energy can be given. The fast yield data included in ENDF/B-IV are defined for a reactor spectrum having an average energy of 0.5 MeV.
- The recent results of Maeck et al (see RP 5) are associated with a spectral index $\sigma_f(^{238}U) / \sigma_f(^{235}U)$.
- The Lawrence Livermore Laboratory (USA) defines a fission-spectrum source as one that gives ^{115g}Cd R-value of 2.80 for ^{235}U fission (i.e. ^{115g}Cd yield ratio from fission spectrum/thermal ^{235}U fission). This definition can be extended to characterize any neutron spectrum or discrete neutron energy.

Some of the measurements in the USA (Maeck) described above together with yield measurements on ^{239}Pu and ^{238}U with monoenergetic fast neutrons in the UK (Cuninghame) should throw light on the problem of the energy dependence of fission yields. It is to be hoped that some suitable parameters by which this effect can be characterized will emerge from these results.

The requirements summarized in Appendix A1 are the ones essential for the nuclear fuel cycle of thermal and fast reactors. The Panel wishes to stress that other fission yield work at neutron energies up to 14 MeV, including measurements in the ^{252}Cf fission spectrum, would not only help in the evaluation of the neutron energy dependence of the fission yields, but would also be essential to improve our understanding of the fission process itself.

(iv) Evaluation work

In view of the increasing volume of experimental data on energy-dependent fission yields it is desirable to rationalize the compilation of these data. It is recommended that evaluators exchange compilations of experimental data with the object of establishing a common computerized data base which can be used by all evaluators and others interested in FPND.

Discrepancies have been discussed in review paper 11a and during the meeting which urgently need clarification. In evaluations in which the normalization procedure (see RP 11a) is used, any changes in some adopted yields would entail a renormalization of the whole yield

curve. This, in turn, involves changes in all FP yields that could occasionally exceed previously assigned uncertainties.

Evalution work is important for the quantitative determination of the dependence of fission yields on incident neutron energy. Evaluators are asked to systematically compare and analyze experimental results from different well defined fast reactor spectra. In parallel, the analysis should be extended to yield data from thermal neutrons to 14 MeV. These investigations should aim at deriving the most suitable and simple description of fast reactor spectra, and presenting the energy dependence of fission yields. Appendix A1 indicates the work required to satisfy user needs.

4.1.3. Recommendations

(i) In the case of thermal fission yields the Panel recommends that evaluators be supported by the pertinent authorities and by the authors of published data in their task to carefully analyze existing experimental data and uncertainties in order to resolve existing discrepancies. In order to reach required accuracies gamma-spectrometric measurements, as recommended in RP 11a, would be sufficient.

(ii) In these cases where the accuracy achieved for fast fission yields (see Appendix A1) does not meet the requirements, further work should be done to reach this accuracy. It is possible, and in many cases probable, that the work in progress summarized above will satisfy the requirements after it has been reported and evaluated.

(iii) Measurers should include in publications specifications of neutron spectra from which they obtain fast fission yields. Work in progress should be completed and reported as soon as possible.

(iv) Investigations of the effect of neutron energy on fission yields should be done by both measurers and evaluators. The present requirements are given in Appendix A1. In order to help establishing the systematics of this effect, the Panel recommends that laboratories be encouraged to continue FP yield studies for incident neutron energies up to 14 MeV or neutron energy spectra other than those of fast reactors (e.g. ^{252}Cf fission neutrons).

(v) Evaluators should aim at establishing, and working from, a common computerized experimental data base.

4.2. Direct and cumulative yields

This section deals with FP yields that cannot be calculated with confidence from total chain yields.

4.2.1. Observations and conclusions

(i) Direct and cumulative yields are needed wherever an inventory of (short-lived) radioactive species is required. Presently needs were expressed for FP release, contamination of reactor components, fuel failure detection, (RP 4) as well as for fuel handling (RP 7). As discussed in section 3.2, the requirements for uncertainties of cumulative yields can be raised to about 30-40% in the case of FP release and contamination of reactor components. Again, the other needs are subject to the limitations expressed in section 3.2 on inventory data.

(ii) FP decay heat: Using mainly calculated (E.A.C. Crouch, AERE-R7680, 1974) and some experimental (Appendix A2, RP 11a) data. the following distribution of fractional cumulative yields of FP listed by C. Devillers for cooling times of 1 s - 100 s (see section 3.3. and Appendix A5) is obtained:

- about 60% of all listed FP, ~70% at 100 s and ~70% of those FP which contribute > 2% to the total FP decay heat have ≥ 95% of the total chain yield.
- only about 20% of all listed FP at 1 s cooling time and 4% at 100 s have ≤ 60% of the total chain yield.
- none of the FP contributing > 2% to the total FP decay heat have < 80% of the chain yield.

This survey is for thermal fission yields of ^{235}U and ^{239}Pu. The situation is not much different for fast fission of these isotopes. The main uncertainties are in calculated fractional independent yields and tend to cancel for cumulative yields of FP with Z > Zp, particularly for cumulative yields close to total chain yields. Therefore calculated cumulative yields of ≥95% should generally be correct within 5%, uncertainties of calculated cumulative yields ≥60-70% should still be within 10-50%, but may be a factor of 2 or more below about 60-70%.

Since a large fraction of the FP contributing to the total decay heat at cooling times up to 100 seconds have low yield uncertainties, and the Panel concluded that rather large uncertainties of individual FPND can be tolerated in this range of cooling times, calculated fractional yields may be sufficient for the time being, until more detailed investigations on needs are available.

(iii) Future needs: Detailed lists of independent and cumulative yields and their accuracies required for FP-decay heat calculations and environmental aspects should be available at the follow-up meeting.

(iv) RP 11a, appendix B, shows that only very few measurements of direct and cumulative yields from thermal neutron fission exist, the majority being for ^{235}U. As Appendix A2 shows, they are just sufficient to fulfil most of the requirements expressed at this Panel. However, the overall uncertainties of the available data should be evaluated, as some remarkable discrepancies are evident in table B1 of RP 11a, and experimental data are often not consistent with a Gaussian charge dispersion curve (cf. Appendix A2).

(v) There are practically no measurements of direct or cumulative yields in fast neutron fission. Much experimental work remains to be done to satisfy the user needs.

(vi) A number of cumulative yield requirements for fuel handling concern branching ratios to metastable and ground states of FP. Since these yields cannot be derived from charge dispersion curves, they have to be measured directly or to be determined with the aid of chain yields and branching ratios.

(vii) The Panel noted that calculational methods for fractional yields using semi-empirical charge dispersion models could help to fill gaps in experimental data. However, experimental data are too scarce to check the general applicability of these models. Furthermore, there is experimental evidence that the presently adopted charge dispersion model, using a unique Gaussian width parameter, is not adequate (see RP 16). Therefore it is at present impossible to estimate meaningful uncertainties in calculated independent yields. A possible exception is ^{235}U thermal fission, where sufficient experimental data are available for deriving Gaussian charge dispersion

parameters of individual mass chains. Further measurements of independent yields are required

- to check the reliability of existing models and help to develop improved models;
- to allow an estimation of the uncertainties of predicted yields;
- to allow a more reliable extension of these models to other heavy nuclides and other non-thermal neutron energies.

RP 16 shows that for the development of improved semi-empirical models further investigations in other areas, which are beyond the scope of this Panel, are also required, such as prompt neutron emission, primary fragment mass and charge, fission theory, etc.

(viii) It should be noted that extensive measurements of direct yields are being performed by Amiel (Israel), the results of which should throw light on charge dispersion models.

4.2.2. Recommendations

(i) Further measurements of independent yields, particularly in ^{235}U thermal fission should be encouraged to improve and check calculational models.

(ii) Measurements of independent FP yields for other heavy nuclides should be initiated to fill the gaps and enable to check predicted yields.

4.3. Calculational Methods

4.3.1. Observations and conclusions

The Panel noted that a number of fission product chain and independent yields required for applications are only available as calculated or estimated values, based on semi-empirical methods and interpolations. The Panel felt that calculated yield curves presented in RP 16 reproduce experimental data well enough to satisfy some user requirements for low accuracy yield data. However, at present uncertainties of calculated yields have only been estimated or obtained for overall fits to experimental mass yield curves. The Panel suggests that empirical uncertainties be derived systematically from a comparison of individual calculated and experimental yield data.

4.3.2. Recommendations

The Panel therefore recommends that

(i) promising work on the development of calculational methods for independent and cumulative FP yields, as reported in RP 16, be further persued;

(ii) semi-empirical models be improved whenever new data become available;

(iii) attempts be made to assign uncertainties to calculated FP yields.

5. DECAY DATA

5.1. Communication of new results

Decay data of short-lived fission products have recently gained increasing importance in applications. Concurrently much effort is being devoted to the measurement of these data and the Panel notes an urgent need that such data reach evaluators and users as fast as possible. Apart from the benefit of the newsletters proposed in chapter 2 the Panel wishes to emphasize that the Recent Reference sections of "Nuclear Data B" are invaluable.

5.2. Observations and conclusions

5.2.1 User requirements

(i) There was some discussion on precision data requirements for environmental research and routine work, but no agreement could be reached. Since no specific data requirements have yet been assessed in this area, the Panel concludes that requests for further measurements should await more detailed sensitivity studies. However, the more general request formulated in Appendix A3 seems to be justified at present.

(ii) The accuracy of half lives of ^{135}I and ^{135}Xe required for reactor kinetics (RP 3) is met.

(iii) Accuracy requirements for decay data put forward in RP 4 for burst fuel detection, FP release and contamination of reactor components are based on general considerations without taking into account available accuracies of FPND. As these requirements are not very stringent,

they are accepted by the Panel and are listed in Appendix A3. Apart from those decay data, for which accuracies have not been assessed, the needs in these areas are either fulfilled or easy and inexpensive to achieve. A revision might be necessary in cases where requirements for FPND other than decay data are not met, based on sensitivity studies.

In quoting accuracy requirements for branching ratios it is assumed that they are the same as those for γ-ray intensities.

(iv) Requirements for FP decay heat can only be given qualitatively at present (see section 3.3). The decay data required for the calculation of the energy released by FP and deposited within the reactor are FP half-lives and effective Q values (Q_{eff}, i.e. $Q-E_\gamma$).

Apart from completely unknown FP, a number of FP with very uncertain half-lives and unknown Q-values contributes to the decay heat at short cooling times up to 100 sec. Although accuracy requirements cannot yet be given, an improvement of the present situation is definitely necessary.

Decay properties of FP important at cooling times $\gtrsim$1000 sec are known, but partly too unprecise to allow a reliable calculation of the partition of the energy released among β, γ and $\bar{\nu}$ (antineutrino).

(v) After some discussion the Panel agreed on burnup requirements as presented in Appendix A3. These include also requirements for non-destructive analysis of spent fuel in safeguards, which have, however, lower priority. The capability and practical applicability of the use of FP activity ratios is still under investigation and may justify needs for more accurate decay data.
The needs for fresh fuel assay in safeguards are agreed by the Panel, especially in view of their low priority.

(vi) Decay data requirements for fuel handling are generally acceptable at present. The Panel concluded that for problems concerning heat released by fission products in spent fuel the knowledge of average β energies in addition to γ-ray data (as proposed for FPND libraries: see annex 3 to chapter 2) are sufficient. However, for shielding (see (viii) below) high energy γ's and β's (bremsstrahlung) are important and the knowledge of their energies and intensities is required.

(vii) Applications in industry, agriculture and life sciences require mainly decay data. The Panel noted that in these fields not only pure FPND are required, but that also other uncertainties discussed in RP 8 and 9

constitute a problem. Nevertheless, FPND of higher accuracy are required for research work and included in Appendix A3.
The Panel supports the request for inclusion of Auger electron data in evaluations.

(viii) A.J. Fudge reported to the Panel that he had observed high energy gamma rays penetrating through thick shields around spent fuel during transport and reprocessing. Several of these gamma rays could not be identifie with the aid of tabulations of known FP gamma ray data, or their abundanc is very uncertain. The Panel endorses the conclusion of Fudge that the observed gamma rays are of very low abundance and not measured in decay property studies. In view of the hazard that high energy gamma rays penetrating through thick shields constitute, efforts should be made to identify and measure them as outlined in Appendix A3.

(ix) Fission yields can be measured rather accurately and inexpensively by gamma spectrometry. Decay data needs depend on the accuracy to which the yield is required. If yields are to be measured to 1-2% accuracy, then half lives and absolute γ-ray intensities are needed to $\pm$ 1% and better.

(x) Prompt γ-rays in fission as well as high energy gamma rays from fission products and bremsstrahlung are sources of photoneutrons. Short-lived FP with high Q values should be studied for high energy γ's and β's. This subject was, however, not discussed by the Panel in greater detail and further investigations on existing information and pertinent FP are still necessary.

5.2.2. Individual decay data

(i) Equilibrium activities: In the case of FP where the daughter reaches equilibrium with its parent rapidly, it is assumed that the equilibrium activity is important for users. The half life of the daughter and the branching ratio of the parent to the daughter are considered to be unimportant in those cases. Instead, radiation intensities of the daughter nuclei per decay of the parent nuclei and their accuracies are included in this survey.

(ii) Half life requirements are generally met at present with the few exceptions noted in Appendix A3. In some cases it would be sufficient to assess the uncertainties or to resolve discrepancies. In future half-lives may be requested for a number of shorter lived FP after needs for environmental aspects, fresh fuel assay in safeguards and afterheat are assessed. The uncertainties of these data, including those of FP listed in Appendix A5, should be evaluated for the next survey of the field of FPND.

(iii) For branching ratios it is assumed that in most fields the required accuracy is expressed as % uncertainty per decay, except if the uncertainty of the branching ratio itself is specifically requested. With these assumptions specifically expressed requirements (Appendix A3) are met.

(iv) A number of needs for absolute gamma ray intensities is not yet met and new measurements are needed. Particularly in gamma ray intensity measurements statistical counting uncertainty and systematic errors (calibration, impurities) should be separated. In addition to the more general recommendations included in chapter 1, the Panel strongly recommends that this be taken into account by evaluators and measurers, and that the latter include pertinent information in their publications. It should be mentioned that for the evaluation of absolute intensities of gamma rays auxiliary decay data like internal conversion electron intensities or conversion coefficients and ground state beta decay branching are required, the accuracy depending on their contribution to the overall error.

(v) Gamma ray energies are generally needed only for identification of FP. The accuracy required has to be compatible with the resolution of commonly used Ge(Li) detectors. Since detectors used in research measurements on gamma rays are usually of higher resolution than those used in application fields, gamma ray energies are sufficiently accurately known, with the exceptions noted in Appendix A3.
Needs for group and mean energies of gamma rays, which are required with much lower accuracy, are a forteriori satisfied.

(vi) β-ray and conversion electron data uncertainties have only been evaluated by Martin and Blichert-Toft (1970) and by Martin (1973) (references [1] and [8] in Appendix A3). However, these evaluations do not include all FP listed in the table of Appendix A3, and the status field had to be left blank for some FP. An inspection of this table

shows that for those FP, for which uncertainties are available, most requirements have been fulfilled. Further requirements for β-ray data not expressed for individual FP arise from evaluations of absolute γ-ray intensities and decay branching ratios, from decay heat calculations and environmental studies.

(vii) It has been noted in subsection 5.2.1 that data on the energy release of individual FP needed for FP decay heat calculations are lacking or inconsistent. In order to deduce effective decay energies from Q values, the energy carried away by antineutrinos ($\bar{E}_\nu$) has to be known, which requires measurements of β-spectra. G. Rudstam informed the Panel that several of the "unknown" FP (see subsection 5.2.1) have been identified at Studsvik, Sweden, and that his laboratory would be in a position to directly measure $\bar{E}_\beta$ and $\bar{E}_\gamma$ separately. Such measurements would be very valuable not only in reducing the number of FP with poorly known decay properties but also would they allow a comparison with data deduced from β-spectra. Upon the recommendation of the Panel these measurements have already been initiated.

(viii) Auger electron and X-ray data are only given by Martin and Blichert-Toft (1970) and Martin (1973) among the evaluations discussed in RP 12 (references [1] and [8] in Appendix A3). The accuracies of available data listed in these publications are presently sufficient to satisfy requirements for applications discussed in RP 8, but do not include all FP for which the data are needed.

5.3. Recommendations

(i) User requirements: further investigations of needs should be performed to yield more specific requirements for individual decay data. This applies to all user areas where needs for individual FP have not yet been assessed, and to those requirements which are based on more general considerations and are not satisfied by available data.

(ii) Evaluations: the survey of the status of decay data presented in Appendix A3 is essentially based on evaluated uncertainties of references [1,2,2a,8] (reference numbers are those of Appendix A3) and is therefore incomplete. The situation is better in the case of half life data (Appendices A3 and A5) where evaluated uncertainties are also available from RP 12 and Nuclear Data Sheets. Therefore the Panel wishes to impress especially on evaluators of decay data the need to give uncertainties, list experimental data and auxiliary assumptions

used in evaluations. Decay schemes and β-branching intensities based on γ-ray measurements and theoretical conversion coefficients should be checked carefully. Data on average β energies, Auger electrons and X rays and their uncertainties should be included in evaluations. An extensive literature search should be made for the next meeting of this kind and lists of further measurements required should be given where data are lacking or unresolved discrepancies exist.

(iii) FP decay heat: the Panel recommends that average β and γ energies be measured for FP where the decay energy is poorly known and supports the initiation of such measurements at Studsvik (Sweden). Furthermore, efforts should be concentrated on measuring β-spectra which have (so far) been studied far less than γ-spectra. A suitable method would be to measure the gross β-spectrum (from low energy to maximum) for individual FP.

(iv) In all cases, where specific needs for individual FP and groups of FP listed in Appendix A3 have clearly not yet been fulfilled (particularly γ-ray intensities), further measurements should be performed to satisfy them.

(v) More emphasis should be given to measurements of intensities of β-transitions to ground states and metastable states with half-lives of more than a few hours (e.g. ^{95m}Nb, ^{99m}Tc). More accurate data, also on conversion electrons, than presently available are required to enable evaluation of absolute γ-ray intensities in those cases where requirements have not yet been met (cf. Appendix A3). For some FP nuclides with very high intensities of ground state β transitions (e.g. ^{144}Ce-^{144}Pr) direct measurement of the gamma ray emission probability is preferable (cf. e.g. the method proposed by K. Debertin, contribution to RP 12, these Panel proceedings, Vol. 3).

6. DELAYED NEUTRONS

Observations and conclusions

6.1. Absolute total delayed neutron yields

The situation for thermal and fission spectrum total delayed neutron yields is considered to be satisfactory for all fissile nuclides in the context of reactor design and operation (review papers 3 and 4). For ^{238}U, where the difference between the older work of Keepin and the new Los Alamos work (table 1, paper 13) is about 15%, the fission spectrum total delayed neutron yield requires checking in order to meet the ±5% accuracy required for the special reactor kinetics purposes outlined in RP 3.

For ^{239}Pu it is observed that the accuracy of the fission spectrum average value of the total delayed neutron yield is only ±8% and does therefore not meet the required ±5%.

The Panel believed that in general the requirements for burst fuel detection (review paper 4) are satisfied by the present data. However, it might be possible that the use of total delayed neutrons for burst fuel detection may be subject to errors due to changes in the fuel composition with time and to diffusion and absorption processes which differentiate between precursor chemical species.

In order to remove systematic errors in delayed neutron yield measurements, which may exceed ±5%, due to differences in normalization, standard neutron sources should be prepared for the calibration of the efficiency of neutron counting equipment. An intercomparison of standard neutron sources used at different laboratories would be required.

6.2. Composite half-life groups

The use of 6 groups seems to satisfy practical requirements. If any need for higher accuracy arises, a splitting into more groups may be necessary alongside more accurate information on the decay curve.

It seems that the data for the group half lives and yields given for thermal and fast fission show only small differences which are probably not significant when considering various fast reactor spectra.

6.3. Delayed neutron precursors

For most practical purposes the chemical and isotopic identity of delayed neutron sources is unimportant except where chemical processes may be of interest: for example, in homogeneous reactor fuels such as in the molten salt fast reactor or in burst fuel detection.

If chemical processes take place one would require better data on delayed neutron yields, delayed neutron emission probabilities (P_n) and fission yields to allow calculations for various situations of fuel composition and chemical fractionations. Fractionation can result in changes of composite half-lives and delayed neutron spectra which may be of interest in such situations.

There are conflicting data at present such that errors up to ±50% exist in these values.

Accurate P_n values and fission yields would allow calculation of total delayed neutron yields for different fuels; alternatively delayed neutron precursor yield measurements for each individual fissionable isotope would be necessary.

Looking further ahead, a consistent set of delayed neutron precursor data is necessary to describe all occurring situations of fuel composition, chemical fractionation and neutron spectrum.

The panel noted that precision measurements of P_n values are in progress in Israel (Amiel) and Sweden (Rudstam).

6.4. Energy spectra

The neutron energy spectrum for delayed neutrons as one group is available but no complete time dependent spectra.

Ifsynthesis of neutron spectra for various fuels or for situations of chemical fractionation is necessary it will be required to measure neutron spectra of individual precursors or at least of time dependent groups.

The Panel noted that G. Rudstam (Studsvik, Sweden) is measuring energy spectra of the following delayed neutron precursors:

(Zn or Ga)-79, Ga-80, Ga-81, Br-87, Br-88, Br-89, Br-90, Br-91, Rb-93, Rb-94, Rb-95, In-129, In-130, (Sn-Sb)-134, Sb-135, Te-136, I-137, I-138, I-139, I-140, (I+Cs)-141, (I+Cs)-142, Cs-143 and Cs-144. By the time of the publication of these Panel proceedings some of the results are already published, others are being prepared for publication.

Recommendations

(i) Further measurements are recommended in order to reach the accuracies required for the fission spectrum averaged total delayed neutron yields for ^{238}U and ^{239}Pu.

(ii) It is recommended that a standard neutron source with an energy spectrum similar to that of total delayed neutrons be prepared for intercomparison of individual laboratory standards used for calibration of neutron counting efficiency.

(iii) The Panel recommends further investigation of the significance of changes in effective delayed neutron data caused by chemical processes in the fuel. This would enable the identification of additional needs for individual delayed neutron precursor data including neutron spectra in the light of the present knowledge and of the measurements yet to be completed.

7. NEUTRON CROSS-SECTIONS

7.1. General recommendations for quoting accuracy requirements

(i) The Panel agreed that for reactor applications target accuracies for neutron reaction cross-sections of FP should be given in absolute values (i.e. b or mb). This is particularly important in cases where a cross-section is unknown in part of- or the whole energy range, since the significance of a particular FP depends upon its reaction rate which is proportional to the product of its reaction cross-section and its yield. The competition between neutron absorption and radioactive decay should also be considered

and the FP classified for accuracy requirements as proposed in RP 10.

(ii) As an exception target accuracies can be given in % if the amount of a product built up by neutron capture in a FP is of primary interest for specific applications such as flux monitoring. In this case the uncertainty in the rate of formation is directly proportional to the relative uncertainty of the capture cross-section irrespective of its value. The most important examples are ^{110m}Ag, $^{134,136}Cs$ and ^{154}Eu formed by neutron capture in ^{109}Ag, $^{133,135}Cs$ and ^{153}Eu, respectively (see Table A4-I).

(iii) The exception mentioned under (ii) above is, however, not applicable for neutron cross-sections as a function of neutron energy, unless restrictions with respect to the magnitude of the point cross-section and range of energy is given. It is recommended to request accuracies for integral quantities (e.g. resonance integral or fast reactor spectrum average cross-section), in % and convert the accuracy of this integral quantity to the absolute accuracy of $\sigma_c(E)$ by the prescripton $\sigma_c(E) \times \phi(E) = \text{const.}$

7.2. Observations and conclusions: user requirements

7.2.1. Physics design of power reactor cores (RP 3)

(i) In order to be able to predict the reactivity effect due to FP in thermal reactors within 2% accuracy, the required accuracies in the capture cross-sections can be expressed as in Table A4-I of Appendix A4. In order to evaluate changes in FP absorption caused by changes in moderator temperature, the shapes of the cross-section curves in the thermal range have to be known to the same accuracies particularly for ^{135}Xe and ^{149}Sm.

(ii) The effect of FP capture in a typical fast breeder reactor can be evaluated to the accuracies adopted by the Panel (section 3.2) if the FP capture cross-sections, averaged over the reactor spectra, can be calculated from differential data (point- or group cross-sections) to the accuracies shown in Table A4-II of Appendix A4. These accuracies are sufficient to satisfy requirements for the Na-void effect, if the shape of $\sigma(E)$ between 0.1 and 100 KeV is known.

(iii) For the Doppler coefficient no special needs for cross-sections of fission products are expressed.

7.2.2. Burnup determination and safeguards (RP 5 and 6)

Neutron capture cross sections are requested to accuracies sufficient to allow corrections for buildup and burnout of important FP to $\lesssim 1\%$ of the burnup at maximum burnups of 40 000 - 70 000 MWd/T (or (2.3-4) x 10^{21} nv t).

(i) Sensitivity studies presented in RP 5 show that the unknown thermal neutron capture cross-section of ^{147}Nd has to be known to $\pm$ 10% if it is of the order of ~200-500 b. In accordance with recommendation 7.1 the Panel agreed on a requirement of $\pm$ 30 b for σ_c of ^{147}Nd. This request has highest priority.

(ii) B.F. Rider has evaluated the accuracy of the thermal capture cross-section of ^{141}Pr required to make the error in the burnup determination via ^{148}Nd due to wrong contamination correction $\lesssim 1\%$ at a thermal reactor burnup of ~40 000 MWd/T (or ~2.3 x 10^{21} n v t). With an average ^{148}Nd thermal fission yield of 1.7% the tolerable uncertainty in the "effective yield" of ^{142}Nd (or ^{142}Nd inventory per fission) is ~0.08% per fission, assuming a ratio of ^{142}Nd: ^{148}Nd ≈ 5:1 in natural Nd. With a thermal fission yield of 5.8% (^{235}U) of ^{141}Pr the required uncertainty of σ_c (^{141}Pr) is $\pm$ 2.8 b at this burnup level. Appendix A4 shows that this requirement is fulfilled by available data.

(iii) Among burnup monitors only thermal capture cross-sections of stable Nd isotopes and of ^{143}Pr are significant. Since the half-life of ^{143}Pr is similar to that of ^{147}Nd, the same accuracy requirement for σ_c can be used. The accuracy required for the thermal σ_c of stable Nd isotopes is $\pm$ 2 b at a maximum fluence of about 4 x 10^{21} n vt. Assuming an epithermal index r ≤ 0.3, the corresponding accuracy requirement for the resonance integral (RI) is $\pm$ 6 b.

According to Fig. 5 of RP 5, the fast reactor averaged neutron capture cross-sections of stable and long lived burnup monitors should be known to about $\pm$ 25-50 mb.

(iv) For the calculation of the ^{134}Cs inventory the Panel adopted the accuracy requirements for thermal neutron capture cross-section data presented in Appendix A4. The uncertainties due to neutron capture in ^{133}Xe and ^{133m}Xe should be $\leq 1\%$ of the ^{133}Cs inventory. Accordingly, the term $\sigma_c\phi$ should be known to $\leq 1\%$ of λ ($\lambda >> \sigma_c\phi$) in the case of ^{133}Xe and $\leq 36\%$ of λ in the case of ^{133m}Xe (branching $^{133}I \rightarrow ^{133m}Xe = 2.8\%$). The corresponding cross-section requirements are $\pm$ 150 b for ^{133}Xe and about $\pm 10^4$ b for ^{133m}Xe at a thermal neutron flux of 10^{14} n$\times$sec$^{-1}\times$cm^{-2}. The value for ^{133}Xe calculated this way agrees with a sensitivity study (table III of RP 6): $\pm$ 153 b uncertainty of σ_c (^{133}Xe) result in 1% error in the ^{134}Cs inventory.

(v) The accuracy requirement of the unknown σ_c of ^{153}Sm can be calculated in the same way: if $\lambda >> \sigma_c\phi$, then σ_c should be known to about $\pm 10^3$ b ($\leq 3\%$ of λ) at a thermal flux of 10^{14} n$\times$cm$^{-2}\times$sec^{-1}.

7.2.3. Engineering design and operation of reactors (RP 4)

Regarding cross-section needs in this area the Panel came to the following conclusions which are partly based on the views expressed by C. Devillers (RP 4):

(i) In order to determine the contamination, the capture cross-sections of stable ^{109}Ag, ^{133}Cs and ^{135}Cs and of the radioactive capture products ^{110m}Ag and ^{134}Cs should be known within $\pm$ 20% for the neutron spectrum involved.

(ii) For a thermal neutron flux of $\phi = 10^{14}$ n cm^{-2} sec^{-1} the term $\sigma\phi$ dominates over λ for ^{135}Xe ($\sigma\phi \approx 20 \times \lambda$). In accordance with the discussion on inventory data needed for FP release in section 3.2, item (iii), the thermal neutron capture cross-section of ^{135}Xe has to be known to $\pm$ 5%. Under these conditions the capture cross-sections of other unstable rare gases can be neglected against λ. For failed fuel detection an accuracy of $\pm$ 10% is requested for the thermal neutron capture cross-section of ^{135}Xe.

(iii) Control rod purposes: A 10% accuracy in the capture cross-sections between 10 keV and 1 MeV is required for ^{151}Eu and ^{153}Eu and, with a lower priority, for ^{152}Eu and ^{154}Eu.

(iv) For gas tagging purposes in fast reactors the capture cross-section of stable Xe and Kr isotopes which are, however, not fission products, has to be known to within 10%. These requirements are not included in Appendix A4.

(v) Other FP capture cross-sections are required if they are relevant compared to the decay constant as discussed in section 3.2. However, no specific FP have been identified by the Panel.

7.2.4. Fuel handling

For fuel handling only the capture cross-sections of FP listed ir Appendix A4 are important. The knowledge of spectrum-averaged cross-sections would be sufficient for this application, if their variations with neutron spectrum are within the requested accuracies.

7.3. Neutron capture cross-sections in the thermal and resonance region

7.3.1 Observations and conclusions

(i) The Panel noted that a number of user requirements on FP capture cross-sections for thermal reactors are not satisfied by available data (see Appendix A4). Further measurements are required.

(ii) Generally user requirements are expressed for effective capture cross-sections, averaged over particular reactor spectra, or for Maxwellian averaged cross-sections and resonance integrals. The knowledge of such integral data may be sufficient for some applications, if their changes with different thermal reactor spectra are within the requested uncertainties. Otherwise the capture cross-section has to be known as a function of incident neutron energy, particularly in the resonance region, the accuracy depending on the shape of the cross-section curve and the neutron energy (see also Appendix A4).

(iii) The Panel considers it desirable to describe the low energy cross-sections of FP in terms of resolved resonances. The energy range to be considered should include about 10-20 resonances. A better knowledge of resonance parameters may also improve the accuracy of calculated neutron capture cross-sections in the KeV range

7.3.2 Recommendations

(i) User requirements should be given for capture cross-sections in the thermal and resonance region separately.

(ii) Further measurements are required in order to satisfy all requirements listed in Appendix A4 which have not yet been fulfilled.

(iii) In addition to cross-section measurements, FP neutron resonance experiments should be pursued with an emphasis on the most important radioactive FP. The purpose of these measurements is to enable a description of the low energy cross-sections in terms of resolved resonances and to provide resonance parameters for calculated cross-sections in the KeV range.

7.4. Status of FP capture cross-sections for neutron energies > 1 KeV

7.4.1 Observations and conclusions

(i) The needed accuracy in σ_c for neutron energies above 1 KeV is not fulfilled by the existing microscopic data, except for some mono-isotopic nuclei.

(ii) Some experiments which may satisfy the needs for σ_c of ^{105}Pd, ^{151}Eu and ^{153}Eu (RPI), Zr and Mo isotopes (ORELA) and experiments at FEI Obninsk are in progress or have been recently completed.

7.4.2. Recommendations

(i) Recognizing that theoretical calculations cannot satisfy all the needs the Panel recommends that microscopic capture cross-section measurements on separated isotopes should be pursued for the most important fission product isotopes.

(ii) The Panel wishes to refer to the general recommendation in the introduction on the necessity for physicists to quote systematic and random errors and to emphasize that this has to be done even for the phenomenological parameters which enter into the nuclear

models used for cross-section calculations, in order to be able to give a good estimate of the error in σ_c . The knowledge of this error is needed for the application of the adjustment method to improve σ_c with the help of integral measurements.

(iii) Noting the importance of average level densities to the theoretical prediction of neutron capture, inelastic and other cross-sections of FP nuclides the Panel recommends that this topic be reviewed within a meeting on the state of the art of theoretical prediction of nuclear data or at a separate specialists meeting within the next two years.

7.5. Status of FP (n,n'), (n,2n) and other cross-sections

7.5.1. Observations and conclusions

(i) There are data in the Italian FPND library for 18 FP isotopes and in the Australian FPND library for a large number of FP isotopes. The ENDF/B-III and IV libraries are using the Australian data file for $\sigma_{nn'}$.

(ii) The needs for FP inelastic scattering cross-sections are as follows:

a) For reactor design, inelastic scattering data need to be improved particularly for neutron energies below 1 MeV.

b) For the analysis of reactivity worth measurements in reactor physics experiments the required accuracy for $\sigma_{nn'}$ might be of the order of 30%. Further studies are needed in order to specify the accuracy for each isotope in a particular experiment.

(iii) The inelastic scattering cross-sections can be calculated to sufficient accuracy for reactor design purposes if the level scheme (energy, spin and parity) of the target nucleus is well known. $\sigma_{nn'}$ should be described for each level separately.

7.5.2. Recommendations

(i) The needs for $\sigma_{n,2n}$, $\sigma_{n,p}$ and other threshold reaction cross-sections for fission products should be investigated.

(ii) The Panel recommends that a compilation and evaluation of nuclear level scheme data needed for calculations of $\sigma_{nn'}$ and σ_{c} be undertaken. It was suggested that evaluators of nuclear structure data include in such an evaluation also the most probable spin and parity values and indicate the possibility of missed levels.

7.6. Integral measurements of FPND

7.6.1. General conclusions and recommendations

Integral measurements have proven to be useful and should continue to be used for checking and/or adjusting FP capture cross-sections, provided that the neutron spectrum is well specified. Results on lumped fission products are not always applicable to all kinds of systems.

7.6.2. Thermal systems

(i) Observation and conclusion: There exists only one good measurement on lumped fission products in thermal systems. This experiment gives only values for thermal capture but no data for resonance capture. The accuracy of this experiment hardly satisfies the accuracy requirements set forth in review paper 3, i.e. 2% on reactivity life time or 5% on neutron capture. However, only if significant improvements in accuracies or values for resonance capture can be obtained should new experiments be considered.

7.6.3. Fast systems

Observations and conclusions

(i) From the STEK measurements on lumped fission product samples deviations of 15-25% between experimental and calculated reactivities are observed. Some conclusions should be drawn after the analysis of the ERMINE measurements will have been completed.

(ii) From integral measurements in FRO on separated isotopes some useful conclusions about the quality of evaluated cross-sections sets can be given. More detailed conclusions can be drawn after the completion of the analysis of the measurements in CFRMF, ERMINE, and STEK mentioned in RP 14.

(iii) For integral measurements a good accuracy (i.e. better than 10%) is needed in σ_c over the neutron energy range important for fast reactors for at least one fission product isotope (e.g. ^{127}I or ^{151}Eu) to be used as reference for relating measured reactivity changes to average capture cross-sections in given neutron spectra.

Recommendations

(i) In view of the different energy ranges which are effective in the various assemblies it is strongly recommended to combine the results of FRO, CFRMF, ERMINE and STEK in a comprehensive analysis of the data.

(ii) In view of the fact that many experimental results on the integral measurements of FP are not available at this time, a specialist meeting on integral measurements of neutron cross-sections and their impact on microscopic cross-section data should be organized as soon as possible. In such a meeting special attention should be given to FP.

8. SUMMARY OF IMPORTANT RECOMMENDATIONS*

8.1. General

(i) Users of FPND should perform detailed investigations of their requirements, specifically by sensitivity studies, to achieve a specification of needs for individual FPND and their accuracies. These investigations should take into account available FPND and their accuracies (sections 1.3, 3.2 and 5.3).

* References to chapters and sections of the original recommendations are given in brackets.

(ii) Measurers are requested to publish all details on experimental conditions, corrections applied and error analysis required for an adequate evaluation (section 1.3).

(iii) Evaluation work should continue to be performed at different places and should in future receive stronger support by pertinent authorities. Evaluators should
- publish all pertinent details of their work;
- assess random and systematic errors separately;
- attempt to resolve discrepancies;
- assign uncertainties to their recommended data;
- warn users in cases of unresolved discrepancies;
- recommend further measurements required where data are lacking or discrepancies exist.

(sections 1.3, 4.1, 5.3, 7.4)

(iv) Unsatisfied user requirements: Further measurements should be performed in all cases where user requirements have not yet been satisfied. Specific recommendations are included in chapters, 4,5,6 and 7, based on a comparison of user requirements with the status of FPND in Appendices A1-A5.

(v) A follow-up meeting should be convened in about three years to review the progress in FPND work stimulated by the present meeting with the aim of setting up a more complete request list (chapter 1).

8.2. International cooperation in the exchange and dissemination of FPND information

(all recommendations: chapter 2)

(i) The list of FPND compilations and evaluations, as provided by Valente from NEA/CCDN (RP 1b) to this meeting should be kept up-to-date and published at annual intervals. To determine the distribution of the list the assistance of Panel participants should be solicited as well as of the Members and Liaison Officers of INDC, EANDC and other regional and national nuclear data committees.

(ii) Two international newsletters, one on activities in the field of compilation and evaluation of FPND and one on measurement activities related to FPND, should be developed as soon as possible. These newsletters should contain information on available manpower, names and addresses,

work finished, underway and planned, recent publications and other pertinent information in brief form. Observed discrepancies should be stated and brought to the attention of the INDC Subcommittee on Discrepancies. After approval by INDC this recommendation should be brought to the attention of EANDC and other regional and national data centres, who should also be asked to assist in determining a suitable distribution. Members and Liaison officers of INDC are asked to make sure that the contribution of their countries to the newsletters are provided regularly and on time to the (still to be determined) publishing centres.

(iii) An international request list for FPND should be developed. The requests should be justified by appropriate sensitivity studies.

(iv) Formats: In order to avoid a proliferation of computer formats the Panel recommends that those developed for the ENDF/B library be adopted as standard formats.

(v) Circular to measurers: The Panel recommends to initiate a circular to measurers stating all information on experimental details required by evaluators which should be included in publications of experimental results. In order to determine the contents of this circular, a questionnaire will be sent to evaluators of FPND. The circular will be distributed to measurers and, in addition, be included in every issue of one of the two newsletters proposed above.

8.3. FP decay heat

(all recommendations: section 3.3)

(i) Error bars should be included in FPND libraries and calculations of the uncertainty of the total FP decay heat should be performed.

(ii) Benchmarks: In order to resolve discrepancies between existing measurements the Panel recommends that benchmark experiments on FP decay heat should be performed at different laboratories under similar irradiation conditions employing all suitable measurement methods. Simultaneously, calculations should be performed at pertinent laboratories for the irradiation conditions of the benchmark experiments and a wide range of cooling times. This should enable a check of the input data of FPND libraries and, together with the measurements, serve as a basis for assessing FPND requirements for FP decay heat. Benchmark experiments and calculations should be restricted to studies of ^{235}U thermal fission.

(iii) Information on finished or on-going studies related to afterheat should be communicated to IAEA/NDS for collection and transmission to the then reviewers of this subject for the follow-up meeting.

8.4. FP yield data

(i) Investigations on the effect of neutron energy on fission yields should be performed by measurers and evaluators. These investigations should help in finding suitable parameters by which this effect can be described and in establishing the systematics of the energy dependence of yields (section 4.1).

(ii) Further measurements of independent and cumulative fission yields should be encouraged to improve and check the prediction of yields by calculational methods (section 4.2).

(iii) The development and improvement of calculational methods for the prediction of fission yields should be further pursued. Attempts should be made to assign uncertainties to calculated FP yields (section 4.3).

8.5. FP decay data

(chapter 5)

(i) Beta decay data should be studied more extensively than in the past. Average β energies, β-spectra and β ray intensities, particularly for transitions to ground and metastable states, should be measured.

(ii) The Panel suggests that data on Auger-electrons be included in compilations and evaluations of decay data.

8.6. Delayed neutron data

(i) In order to avoid systematic differences in delayed neutron measurements due to counter-calibration the Panel recommends to prepare a standard neutron source with an energy spectrum similar to that of total delayed neutrons for inter-comparison with individual laboratory standards (chapter 6).

8.7. Neutron cross-section data

(i) Noting the importance of average level densities to the theoretical prediction of neutron capture, inelastic and other cross-sections of FP nuclides the Panel recommends that this topic be reviewed within a meeting on the state of the art of theoretical prediction of nuclear data or at a separate specialists meeting within the next two years (section 7.4).

(ii) The Panel recommends that a compilation and evaluation of nuclear level scheme data needed for calculations of $\sigma_{nn'}$ and $\sigma_{n\gamma}$ be undertaken. It was suggested that evaluators of nuclear structure data include in such an evaluation also the most probable spin and parity values and indicate the possibility of missed levels (section 7.5).

(iii) In view of the different energy ranges which are effective in the various assemblies used for measurements of integral FP cross-sections it is strongly recommended to combine the results of FRO, CFRMF, ERMINE and STEK experiments in a comprehensive analysis of the data.

(iv) In view of the fact that many experimental results on the integral measurements of FP are not available at this time, a specialists meeting on integral measurements of neutron cross-sections and their impact on microscopic cross-section data should be organized as soon as possible. In such a meeting special attention should be given to FP (section 7.6).

Appendix A1: FP CHAIN YIELDS

Comparison: status - user requirements

A1.1 Chain yields from thermal neutron fission

(i) The uncertainties of FP yields from thermal fission are those recommended by Walker (RP 11a). Since the uncertainties of chain yields in the peak regions of the mass yield curve do not differ much, and since most of the requirements are fulfilled, presently available accuracies are presented for all requested mass chains as a group, together with the user needs in Table A1-I.

In some cases the uncertainties of presently available data, and/or discrepancies among experimental data, which cannot be resolved, exceed the accuracy required. These chain yields are indicated under "comments" in Table A1-I and surveyed in more detail in Table A1-II. Discrepancies are discussed in RP 11a, together with measurements required to resolve them and check existing data.

However, it is the feeling of the Panel that the status of thermal chain yields is so close to the required accuracies that an improvement of the data has no high priority (see chapter 4).

(ii) Safeguards: ^{154}Eu, formed from ^{153}Eu by neutron capture with contributions from lower mass chains by multiple capture is used in isotope correlation studies. An improvement of present data is needed, but the definite accuracy required is not yet known. This request has very low priority.

(iii) Fuel design: Requirements for gas pressure calculation are included in Table A1-I. Requirements for information on the chemical state in nuclear fuel are $\pm$ 15% for yields $\geq$ 1%. These requirements are met by presently available thermal chain yields (see RP 11a) with the exception of the few cases where yields have not been measured:

- ^{233}U: 128-130
- ^{235}U: 130
- ^{239}Pu: 107, 108, 129, 130
- ^{241}Pu: 89, 98, 100, 103, 105, 107-110, 130, 139

Table A1-I: Chain yields from thermal fission: status and user requirements.

application field	FP mass numbers [a]	fissioning nuclei	accuracy(%) needs	accuracy(%) status	comment
burnup RP 5	106,133,137,143 144,145,146,148 150	^{233}U	2-3	≤ 3	except A = 106,133
		^{235}U	1-2	≤ 2	except A = 106,133
		^{239}Pu	1-2	≤ 2	except A = 106,133,137
		^{241}Pu	5	< 5	except A = 106
burnup RP 5	95,103,140,141	^{233}U	5	≤ 5	except A = 103,141
		^{235}U	3	≤ 2	except discrepancy
		^{239}Pu	3	2-5	also discrepancies
		^{241}Pu	5	< 5	except A = 103: no measurement
safeguards RP 6	149,151-153	^{235}U	-	~ 6	improvement needed
		^{239}Pu	-	3-10	other requirements: see burnup
fuel handling RP 7	89,90,91,95 103,106,125,129 131,133,137,140 141,143,144,147, 151,153	^{233}U	5-10	≤ 7	except A = 125,129 discrepancies exist, see Table A1-II A=153: status ~ 10%
		^{235}U		≤ 7	
		^{239}Pu		≤ 7	
Physics design of reactor cores RP 3	99,103,131,133 143,147,149, 151,152	^{233}U	6-11	≤ 6-7	requirements met, except discrepancies at A = 103,131, see Table A1-II
		^{235}U		≤ 6-7	
		^{239}Pu		≤ 6-7	
fuel design (gas pressure)	83,84,86,131, 132,134,136	^{233}U	10	<10	
		^{235}U			
		^{239}Pu			
FP release and contamination RP 4	90,95,103,106 109,125,131,133 135,137,140	^{233}U	20	< 20	requirements met; see also cumulative yields Appendix A2.
		^{235}U			
		^{239}Pu			

a) Other mass numbers, for which only cumulative yields are required, are included in appendix A2 (e.g. 135Xe).

Table A1-II: Individual chain yields in thermal fission: unsatisfied requirements

FP mass no	fissile isotope	status (%)	accuracy required a) (%)	RP	P	Comments D = discrepancies among experimental data
95	^{239}Pu	1.6	3	5	2	D of 5% and more: see RP 11a
103	^{233}U	6	5	5	2	D even higher, see RP 11a } RP 3
	^{235}U	1.4	3	5	2	for D see discussion RP 11a } requires
	^{239}Pu	4.2	3	5	2	for D see discussion RP 11a } 6% accuracy
	^{241}Pu	?	5	5	2	no measurements
106	^{233}U	4	2-3	5	2	
	^{235}U	3	1-2	5	2	
	^{239}Pu	5	1-2	5	2	D even higher, see RP 11a
	^{241}Pu	6.6	5	5	2	
125	^{233}U	13	5-10	7	3	based on only one measurement
	^{235}U	7	5-10	7	3	D of 30% and more exist
	^{239}Pu	14	5-10	7	3	
129	^{233}U	?	5-10	7	2	no measurements
	^{235}U	23	5-10	7	2	
	^{239}Pu	?	5-10	7	2	no measurements
131	^{239}Pu	2.4	5-10	7	3	D and inconsistencies exceeding
			8	3	3	required accuracy: see RP 11a
133	^{233}U	3.5	2-3	5	2	D among experimental data, especially
	^{235}U	2.4	1-2	5	2	between Xe and Cs.
	^{239}Pu	2.7	1-2	5	2	^{239}Pu: see discussion RP 11a
137	^{233}U	2.3	2-3	5	2	D among experimental data exist, but
	^{239}Pu	2.7	1-2	5	2	average probably adequate.
141	^{233}U	7.6	5	5	3	
	^{239}Pu	4.7	3	5	3	

a) Only the review paper requesting highest accuracy is listed, except in cases of about equivalent accuracy or if the primary request has low priority.

P = Priority for improvement of present status: 1 = high
2 = medium
3 = low

Since other chain yields are known much more accurately than required, the overall accuracy should be sufficient (see chapter 4.), with the possible exception of ^{241}Pu.

Requirements for yields < 1% ($\pm$ 50% for yields between 0.1% and 1% and within a factor 5 for yields < 0.1%) can be satisfied by interpolated and extrapolated yields supplementing experimental data.

A1.2 On the effect of incident neutron energy on yields

Fission yields in the region of the wings of peaks and in the valley of the mass distribution increase with increasing incident neutron energy. When going from thermal to fast reactor fission, the yields in the valley are enhanced by a factor of 3 for ^{235}U and a factor of 2 for ^{239}Pu.

This increase has to be compensated by a decrease in peak yields as the sum of all yields is 200%. However, the two-mode fission theory predicts that the change in peak yields is $\leq$ 3% from thermal to fast reactor fission. Therefore the variation of peak yields with different fast reactor spectra is expected to be even smaller

A1.3 Chain yields from fast neutron fission

(i) The main survey of data requirements and status presented in Table A1-III, is restricted to chain yields from fast fission of ^{235}U and ^{239}Pu, needed for current fast reactor programmes. The term "fast" used in this survey needs some further explanations (it is also discussed in section 4.2.).

Users require fission yields for fast reactor spectra. Available data have to be valid for the particular fast reactor spectrum involved within the requested accuracy limits.

In current evaluations "fast yields" are not well defined. Recommended yields are derived from experimental results obtained in different fast reactor spectra, fission neutron spectra (generally from ^{235}U, ^{238}U or ^{252}Cf), and sometimes even at discrete neutron energies ranging from 1.5 to 3 MeV. In order to allow a better comparison of this rather poorly defined status with user requirements, a range of values is given in Table A1-III (see explaination below). These values are derived from the survey given in RP 11b and information supplied by Panel participants.

In almost all cases, the lower limit is the status supplied by B.F. Rider. It corresponds to the data presently adopted for the ENDF/B file and published in [1]. It is important to note that weighted averages are dominated by the most accurate results and the uncertainty of the mean is even lower than experimental errors if the data are consistent. Contrary to other evaluators, Meek and Rider have also included estimated values in their evaluation procedure and have derived fast from thermal yields by drawing smooth curves through measured fast/thermal yield ratios in order to get completely evaluated mass yield curves. In very few cases the uncertainty of the most recent results of von Gunten (see section 4.2) are used.

Since the most accurate results were always obtained from measurements in fast reactor spectra, the lower limits represent the status achieved experimentally for particular fast reactor spectra. Uncertainties as low as 1-1.5% indicate that different measurements are fairly consistent and any effect of fast reactor neutron spectrum on yields falls within experimental error.

The upper limit is

- either derived from J.G. Cuninghame's judgement of the status, based on his survey of the subject for the Panel (RP 11b);
- or represents differences between yields recommended by Meek and Rider [1] and Crouch [2]; this partly supersedes the comparison in RP 11b between [2] and the earlier evaluation of Meek and Rider [3];
- or is the uncertainty given by Crouch [2] as the larger standard deviation of weighted or unweighted average.

Thus the upper limit reflects existing discrepancies and/or a possible effect of neutron energy on yields and/or the overall reliability of evaluated yields.

When comparing the status of yields with user requirements, the upper limit should be used (except for very large discrepancies), unless the evaluations or sources of experimental data are consulted directly. Yield data for other fissioning nuclei present in smaller amounts in fast reactor fuel are needed with much lower accuracy. It would be worth investigating if this required accuracy could be achieved using fission theory and systematics of mass yields (see 4.1 and RP 16).

Table A1-III: ^{235}U and ^{239}Pu fission yields from a fast reactor spectrum: status and user requirements

FP mass no.	relative accuracy (%) of fast chain yields								
	status[a]		user requirements[b]						
	^{235}U	^{239}Pu	review paper no.				fuel[c]	dosimetry	
			3	4	5	7	design	US[d]	others
83	2-7	2-8					10		
84	2-4	2-5					10		
85	2-5	2-5		20		5-10*	10*		
86	2-3	2-5					10		
87	2-5	3-6		≤20*					
88	2-4	3-7		≤20*					
89	3-6	3-6		<20*		5-10			
90	2-3	1.5-5		20		5-10			
91	2-10	1.5-6		<20*		5-10			
95	1.5-3	2-3	30	20	≤2	5-10		2	10
97	1.5-3	3-4	30						
98	1.5-3	2-6	35						
99	1.5-5	~3	20			10-20			
100	1.5-4	5-6	35						
101	2-7	2-7	20						
102	2-4	3-7	25						
103	2-6	3-5	20	20	≤2	5-10			
104	2-5	3-7	30						
105	4-26	≥6	20						
106	6-30	3-10	35	20	≤2	5-10			
107	≥16	≥16	25						
109	≥12	6-18	40	20		10-20			
111	2-40	12-25				10-20			
123	≥12	≥16				10-20			
125	≥8	≥8		20		5-10			
127	≥12	≥12		≤20*		10-20			
129	6-12	15		≤20*		5-10			
131	1.5-3	2-10	30	20		5-10	10		
132	1.5-6	3-5		≤20*		10-20	10		
133	1.5-3	2-5	20	20	2	5-10			

Table A1-III: (continued)

FP mass no.	relative accuracy (%) of fast chain yields								
	status[a]		user requirements[b]						
	^{235}U	^{239}Pu	review paper no.				fuel[c]	dosimetry	
			3	4	5	7	design	US[d]	others
134	2-7	4-5					10		
135	2-4	3-6	30	20					
136	1-5	3-5					10		
137	1-6	2-10	40	20	≤2	5-10		2	10
138	1.5-3	4-25		<20*					
139	1-5	6-10	40	<20*					
140	1.5-4	1.5-3		20	≤2	5-10		2	10
141	2-3	3-5	35	<20*	≤2	5-10			
143	1-5	1.5-7	30		≤2	5-10		2	10
144	1.5-6	2-3			≤2	5-10			
145	1-3	1.5-5	35		≤2			2	10
146	1-4	1.5-5			2			2	10
147	3-18	3-6	25			5-10			
148	1-3	1.5-6			≤2			2	10
149	3-5	2-5	30			10-20			
150	1.5-5	1.5-5			≤2			2	10
151	2-3	3-5	25			5-10			
152	2-3	3-6	55						
153	≥8	≥12	40			5-10			
155	12-50	≥16	55			5-10			
156	8-60	≥6				10-20			

a) For references and discussion of lower/upper limit of status see text.

b) In the user fields discussed in review papers 3, 4 and 7 and in fuel design, requirements for yields from ^{232}Th, $^{233,238}U$ and $^{240,241,242}Pu$ fast fission are not listed separately. They are about a factor of 2-5 less stringent than those for ^{235}U and ^{239}Pu.

* .. chain yield required for calculation of cumulative yield

c) Requirements listed are only for evaluation of gas pressure within fuel.

d) The accuracy listed is only required by the LMFBR and FFTR programme at Hanford (USA).

(ii) As burnup determination and fast reactor dosimetry require yields from fast fission of nuclei other than ^{235}U and ^{239}Pu to much higher accuracy than for other applications, their needs are listed separately in Table A1-IV together with the data status.

Only calculated fast yields together with their estimated uncertainties are available for ^{236}U, $^{240,241,242}Pu$ and $^{241,243}Am$. Peak yields for ^{241}Pu fast fission can be derived from thermal yields more reliably than for the other nuclides listed above; the estimated uncertainty limits are therefore lower.

The status for ^{233}U and ^{237}Np corresponds to information supplied by Rider and Cuninghame and the evaluation of Crouch [2].

The same sources as well as a contribution by Lammer [4] are used for ^{232}Th and ^{238}U. However, the low uncertainties of Rider (see [1])are not used for most yield data, as unresolved discrepancies make the absolute yields obtained from a normalization uncertain (see detailed discussion in [4]).

(iii) The knowledge of the complete mass yield curve is required for investigations of the chemical state of fast reactor fuel. Of primary importance are all major fast chain yields, i.e. the mass ranges 80-110 and 125-155. As discussed in section 4.1, the requirements expressed by the subgroup on chain yields (SG) and by R.H. Flowers (F) are:

$\pm \leq$ 10%(SG) - 15%(F) for ^{233}U, ^{235}U, ^{239}Pu and ^{241}Pu
$\pm$ 15%(F) for ^{232}Th, ^{238}U and ^{240}Pu

These chain yields have to be known also for the calculation of afterheat.

Requirements for yields at the wings of the peaks and in the valley of mass distributions as expressed by Flowers (section 3.2) for the fissionable nuclei listed above are:

$\pm$ 50% for chain yields between 0.1 and 1%
within a factor 5 for chain yields < 0.1%.

(iv) For particular fast reactor spectra (corresponding to the lower limit in Table A1-III) many requirements for burnup and dosimetry applications have been fulfilled for yields from ^{235}U and ^{239}Pu fission. However, the data have to be analyzed in more detail for appli-

Table A1-IV: Chain yields from fast fission of other nuclei: status and requirements

mass no.	needed for[a]	status: relative uncertainty (%) of available data[b]									
		^{232}Th	^{233}U	^{236}U	^{238}U	^{237}Np	^{240}Pu	^{241}Pu	^{242}Pu	^{241}Am	^{243}Am
95	b,d	6-11	10	↑	3	6	↑	↑	↑	↑	↑
103	b	6-10	10-15		3-4	10					
106	b	10-25	6-15		10	10					
133	b	6-20	10		8-20	10					
137	b,d	6-25	5-10		6-20	5-10					
140	b,d	6-12	8-10		2	3-4					
141	b	6	8-15	15-20*	8	20	15-20*	6-15*	15-20*	15-20*	15-20*
143	b,d	8	10		8	10					
144	b	5-10	10		8	4					
145	b,d	8	10		8	15*					
146	b,d	10	10-15*		8	15*					
148	b,d	8	10-15*		8	9					
150	b,d	10	10-15*	↓	8	15*	↓	↓	↓	↓	↓
purpose		accuracy (%) required									
burnup		≤5	≤2	-	≤5	≤10	≤10	≤3	≤10	≤10	≤10
dosimetry[c]		2	-	2	2	2	-	-	-	-	-

a) b ... burnup, d ... dosimetry.

b) Status: for references and discussion of lower/upper limit see text.

c) These requirements are only for the LMFBR and FTR programme at Hanford, US. Other requirements are ± 10%.

* Only available as calculated yields (Sidebotham, see RP 11b and 16), estimated uncertainty.

cation and the effect of neutron energy on fast yields has to be considered (see below). The bulk of other burnup requirements is clearly not fulfilled.

Other user requirements for ^{235}U and ^{239}Pu fast yields are generally fulfilled, particularly if a different allocation of accuracy targets using available yield uncertainties is considered. Available fast yields from other nuclei except ^{232}Th and ^{238}U are not sufficient to satisfy needs for investigations of the chemical state of nuclear fuel.

An improvement of the situation is to be expected after the completion of current measurements listed in section 4.1.2. But additional evaluation work is required, as complete mass yield curves are not being investigated experimentally.

A1.4 Effect of neutron energy on yields

(i) Generally speaking, changes of FP chain yields as a function of fast reactor neutron spectrum should be known within the accuracy limits given in Table A1-III.

(ii) Burnup: At present the most widely used burnup monitor is ^{148}Nd, measured mass spectrometrically together with other Nd isotopes. It has been observed (RP 5) that relative Nd yields change systematically with fast reactor neutron spectrum. This change has to be known for ^{232}Th, ^{233}, ^{235}, ^{238}U and ^{239}, ^{241}Pu with the accuracies listed in Tables A1-III and A1-IV.

(iii) Apart from a recent study on ^{235}U fast yields presented to the Panel by W.J. Maeck (see also RP 5) no data are available on the variation of fission yields in the energy range of interest for fast reactors.

Measurements to $\pm$ 2% accuracy on the variation of Nd yields with neutron energy or fast reactor spectrum are required to enable the selection of most suitable burnup monitor nuclides. Such measurements should also include other nuclides in the mass range 140-160 to help in establishing the systematics of the energy effect. Further measurements with lower accuracy are needed for yields at wings and in the valley region to establish the energy effect for the chain yields requested in Table A1-III.

References:

[1] M.E. Meek and B.F. Rider, NEDO-12154-1 (Jan.1974)

[2] E.A.C. Crouch, AERE-R7394

[3] M.E. Meek and B.F. Rider, NEDO-12154 (Jan.1972)

[4] M. Lammer, contribution to RP 11b, these Panel proceedings, Vol. 3.

Appendix A2: INDEPENDENT AND CUMULATIVE FISSION YIELDS

Comparison: status - user requirements

A2.1 Present and future needs

The present survey includes only those needs which were expressed explicitly by the Panel. It should be noted, however, that this survey represents only a minority of requirements for independent and cumulative yield data. Many more FP will have to be included as soon as needs for afterheat and environmental aspects are assessed in detail.

A2.2 Cumulative yields in thermal neutron fission

(i) The status of cumulative yields from ^{233}U, ^{235}U and ^{239}Pu thermal fission is compared to user requirements in Table A2-I. An inspection of this table shows that most of the present requirements for cumulative yields in thermal neutron fission are met. This is mainly due to the fact that the majority of needs are expressed for the Kr and Xe isotopes which have been measured more extensively than other FP. Few data are available for ^{241}Pu (see below), and none have been requested. If they are required, accuracies will probably be a factor of 2-3 less stringent than those listed in Table A2-I.

(ii) Basic data: Fractional cumulative yields and their uncertainties have been evaluated by M. Lammer (as shown in the Annex to Appendix A2), based on the compilation of experimental data by W.H. Walker (RP 11a, Appendix B) and have been approved by Panel participants. Only those mass chains have been evaluated, for which at least one measurement of a fractional yield is available. However, Table A2-I contains the status of only those data which could be derived from measurements with sufficient confidence (see Annex to this appendix). The data status is marked with an asterisk (*) in Table A2-I if the corresponding fractional cumulative yield and its uncertainty was not obtained directly from measurements.

(iii) Uncertainties: Uncertainties of chain yields are those recommended by Walker, RP 11a. They are taken in quadrature with the uncertainties of fractional cumulative yields to calculate the uncertainties of cumulative yields, which are also shown in Table A2-I.

Table A2-I: Cumulative yields in thermal fission: status and requirements

FP	uncertainty (%)[a] for fission of			accuracy (%) required for[b] R P 4		others
	^{235}U	^{233}U	^{239}Pu	rc	ff	
T	~10	~10	~12	50		5-10 (RP7)
^{85m}Kr	2.5	3	~3	20	20	
^{85}Kr	2.3	2.3	2.7	20		5-10 (RP7),10(fd)
^{87}Kr	3.4*	?	~3.6*	20	20	
^{88}Kr	4.5*	?	<10*	20	20	
^{89}Kr	2.3	2	2.6	20	20	
^{90}Kr	3	2.1	2.4	20	20	
^{90}Rb	3	3.4*	~4*	20		
^{91}Kr	2.7	3.3	3.6		20	
^{129m}Te	?[c]	?[c]	?[c]	20		
^{133}I	≤2.6	5.3*	5*	20		
^{135}I	2.4	5.6	3.5	20		5(RP3)
^{135}Xe	2.4	2.6	3.5	20	20	5(RP3)
^{137}Xe	2.6	2.6	~3	20		
^{138}Xe	2.5	~3	6.6		20	
^{139}Xe	3.5	~4	5.6		20	
^{140}Xe	3.5	~5	3.6	20	20	
^{140}Cs	2.2	~6	?	20		
^{140}Ba	1	1.9	~10*	20		3-5(RP5)
^{141}Xe	10	~10	~6		20	

Table A2-I: continued

a) - Estimated uncertainties, marked with (*), are included here only if they are well below the requirements. Otherwise they are marked with (?) but included in the Annex to this appendix, except ^{129m}Te: see c).
- For ^{241}Pu see text.

b) rc ..... FP release and contamination of reactor components
ff ..... failed fuel detection
fd ..... fuel design

c) No measurements exist. To be used with ^{129}Sb or mass 129 yield and decay branching.

* estimated

Note: 1) Some of the fractional yields are based on single measurements with low uncertainties. However, not all of these uncertainties are reliable, as some discrepancies among data presented in Table B1 of RP 11a and in the Annex to this appendix show. Furthermore, part of the experimental data of fractional yields and their uncertainties have been derived from published chain yields, which do not correspond to those presented in RP 11a and adopted by the Panel.

2) In most measurements independent and cumulative yields are determined directly. Fractional yields are derived subsequently from known standard yields and chain yields. The uncertainties of these yields are therefore included in the uncertainties of the fractional yields as listed by Walker. Since the latter are again combined with uncertainties of chain yields, the resulting uncertainty of the cumulative yield is higher than that of the original measurement. It is, however, not the intent of this survey to go back to the original publications, and the combined uncertainties are probably more realistic in the light of the arguments under 1) above.

(iv) In ^{241}Pu thermal fission only the cumulative yields for ^{85}Kr and mass 135 are available. The uncertainties, taken from RP 11a (for ^{85}Kr see Annex), are:
^{85}Kr: 2.3%, ^{135}I and ^{135}Xe: 3.4%

A2.3 Cumulative yields in fast neutron fission

The only experimental data available are fractional independent yields of ^{133}I, ^{135}I and ^{135m}Xe in fast fission of ^{232}Th (see RP 11b). Therefore only user requirements are summarized in this survey and presented in Table A2-II . In view of the scarcity of data it is indicated in the table, for which FP, at least within the required accuracy, total chain yields can be used. Clearly, a lot of experimental work is still required to fulfill the needs.

From available experimental data the following comments can be made on the data status:

(i) The measurements of ^{133}I and ^{135}I fractional independent yields in Th-232 fast fission suggest that the limits for fractional cumulative yields given in Table A2-II hold for all fissile nuclides, assuming Gaussian charge dispersion. Using the uncertainties of fast chain yields given in Table A1-II the following uncertainties can be deduced for cumulative yields from ^{235}U and ^{239}Pu fast fission:

$\leq \pm$ 4% for ^{133}I

$< \pm$ 5% for ^{135}I and ^{135}Xe

(ii) The cumulative yield of ^{85}Kr can be deduced from the chain yield given in Table A1-II and the branching $^{85m}Kr \rightarrow {}^{85}Kr$ (see discussion in the Annex). The uncertainties are:

$\leq$ 5% for ^{235}U and ^{239}Pu fission in a typical fast reactor spectrum, and

$\leq$10% taking into account the energy dependence of the mass 85 yield.

Table A2-II: Cumulative yields in fast fission: user requirements

FP	required accuracy (%)[a]			
	RP 4			
	r.c.	f.f.	RP7	comments
T	50		5-10	no data available
^{85m}Kr	20	20		
^{85}Kr	20		5-10	± 10% for fuel design (see text, also for status)
^{87}Kr	20	20		total chain yield can be used for ^{235}U and ^{239}Pu
^{88}Kr	20	20		
^{89}Kr	20	20		
^{90}Kr	20	20		
^{90}Rb	20			
^{91}Kr		20		
^{127m}Te			10-20	can be deduced from chain yield and branching ratio
^{129m}Te	20		10-20	
^{132}Te				
^{133}I	20			$\gtrsim$ .99 of chain yield (see text)
^{135}I	20			$\gtrsim$ .98 of chain yield (see text)
^{135m}Xe	20	20		
^{135}Xe	20	20		≈ total chain yield
^{137}Xe	20	20		
^{138}Xe	20	20		
^{139}Xe	20	20		
^{140}Xe	20			
^{140}Cs	20			
^{141}Xe		20		

a) The accuracies listed are for fast yields of the main fissile isotopes ^{235}U and ^{239}Pu. Requirements for ^{232}Th, $^{233,236,238}U$, $^{240,241,242}Pu$ are a factor of 2-5 less stringent.

r.c. = FP release and contamination of reactor components

f.f. = fuel failure detection.

Annex to Appendix A2:

EVALUATION OF FRACTIONAL YIELDS

by M. Lammer

1. Abbreviations

The abbreviations given below are used throughout the text and the tables.

chy ..... chain yield(s)
cy ..... cumulative yield(s)
e,exp .... experiment(al)
fc ..... fractional cumulative yield(s)
fi ..... fractional independent yield(s)
G ..... (assuming) Gaussian charge dispersion
w,wa ..... weighted average
Zp ..... most probable charge
σ ..... Width of Gaussian charge dispersion

2. ^{85}Kr and ^{85m}Kr

In currently adopted decay schemes there is no direct branch from ^{85}Br to the ground state of ^{85}Kr. The adopted internal transition branch is:

$$^{85m}Kr \longrightarrow {}^{85g}Kr: \quad 0.212 \pm 0.009$$

Lisman et al (J. inorg. nucl. Chem. 33 (1971) 643) measured both ^{85g}Kr and ^{85}Rb by isotope dilution mass spectrometry and obtained fractional cumulative yields for ^{85g}Kr. These yields, together with the differences between them and the branching ratio from the decay of ^{85m}Kr (d) are listed below:

	^{233}U	^{235}U	^{239}Pu	^{241}Pu
fc	0.2296 ± 0.0028	0.2176 ± 0.0024	0.2260 ± 0.0018	0.2235 ± 0.0033
d	0.018	0.006	0.014	0.012

Only the values for ^{235}U and ^{241}Pu agree with the branching ratio within the error limits. For deriving the fractional cumulative yields of ^{85m}Kr shown in Tables A2III-A2V, these differences, rounded upwards, were taken as upper limits of the fractional independent yield of ^{85g}Kr.
Since the yield of ^{85g}Kr can be measured mass spectrometrically relative to stable FP Kr, the uncertainties assigned by Walker (RP 11a) to mass 84 and 86 chain yields are assigned in Table A2-II to the cumulative yield of ^{85g}Kr.

3. Summary of fractional yields

Tables A2-III to A2-V show in detail how the uncertainties listed in Table A2-I were obtained. Only those mass chains are included, for which at least one measurement on fractional yields is available. Wherever possible, fc were deduced directly from measurements. This includes cases where fc of a FP has not been measured directly, but deduced from fi of this FP on the next member in the chain assuming a Gaussian charge dispersion.

The Tables are generally self-explanatory, but some comments are given below. If several measurements of different fi and fc were available for a mass chain, they were plotted on probability paper and evaluated in detail. Experimental data are taken from RP 11a [1].

3.1. Table A2-III: fractional yields in ^{235}U thermal fission

Detailed evaluations of mass chains 87,88,91,139 and 140 are presented in Tables A2-VI to A2-X, and discussed further below. Fractional yields for mass 91 are presented in Table A2-VIII in order to illustrate their consistency with G in spite of their apparent disagreement with the calculated data of Crouch [2].

The selection of all other data in Table A2-III is self-evident.

Table A2-III: Details on status of cumulative yields from ^{235}U thermal fission

FP	fc	uncertainty (%) for			comments and data: underlined are data used to obtain fc e ... experimental, c ... calculated, w ... weighted average
		fc	chy	cy	
^{85m}Kr	≳.99	<1	2.3	2.5	for ^{85m}Kr and ^{85}Kr see text
^{87}Kr	≳.99	2	2.8	3.4	see Table A2-VI
^{88}Kr	.96	4	2	4.5	see Table A2-VII
^{89}Kr	.96	1	2.1	2.3	^{89}Rb fi: e .042 ± .007w, c .04, G / ^{89}Kr fc: c .96
^{90}Kr	.86	2.3	1.9	3	^{90}Rb fc: c .9976; fi: e .13 ± .01, c .15/^{90}Kr fc: e .86 ± .02, c .85;
^{90}Rb	.99	2.3	1.9	3	fi: e .63 ± .08, c .62
^{91}Kr	.59	2	1.8	2.7	see Table A2-VIII
^{133}I	1.00	≤1	2.4	≤2.6	^{133}I fi: e .024 ± .005w, G, used
^{135}I	.965	.4	2.4	2.4	as evaluated by Walker, RP 11a;
^{135}Xe	1.00	-	2.4	2.4	for ^{135m}Xe the decay branch from ^{135}I should be used.
^{137}Xe	.978	.3	2.6	2.6	^{137}Xe fc: e .978 ± .003
^{138}Xe	.952	.1	2.5	2.5	^{138}Cs fi: e .048 ± .001w, used with G
^{139}Xe	.80	3	1.8	3.5	see Table A2-X
^{140}Xe	.60	3.3	1	3.5	evaluated: Ba: fi = .07 ± .02, Cs: fi = .33 ± .02
^{140}Cs	.93	2	1	2.2	^{141}Xe fc: e .21 ± .02, fi: .20 ± .03
^{141}Xe	.21	10	1	10	^{141}Xe fc: e .21 ± .02, fi: .20 ± .03

Table A2-IV: Details on status of cumulative yields from ^{233}U thermal fission

FP	fc	uncertainty(%) for			comments and data: data used to obtain fc are underlined e ... experimental, c ... calculated, w ... weighted average
		fc	chy	cy	
^{85m}Kr	$\gtrsim$.98	~2	2.3	3	for ^{85m}Kr and ^{85}Kr see text
^{87}Kr	~.88*	~20*	2.3	20	^{87}Kr fc: c.98 / ^{87}Br fc: e.56±.04, c.68 ; fi: e.75±.20, c.56/ ^{87}Se fc: e.19±.04, c .12 / G (see text)
^{88}Kr	$\gtrsim$.80*	~25*	1.6	25	^{88}Kr fc: c.91/ ^{88}Br fc: e.47±.06, c.38; G, very uncertain (see text)
^{89}Kr	.86	1	1.6	2	^{89}Kr fc: e .86 ± .01, (c.88)
^{90}Kr	.67	1.5	1.5	2.1	^{90}Y fi: e .00008, c .0004/ ^{90}Rb fc: c .96 / ^{90}Kr fc: e .67±.01, c.32/ ^{90}Br fc: e .10±.03, c .053 / G, all e data used (see text)
^{90}Rb	~ .98*	~3*	1.5	3.4	
^{91}Kr	.33	3	1.2	3.3	^{91}Kr fc: e .33±.01
^{133}I	$\gtrsim$.98*	~4 *	3.5	$\lesssim$5.3	^{133}I fc: c .999; fi: e .14±.01, .155±.010, c .17/G (see text)
^{135}I	.79	5	2.4	5.6	as evaluated by Walker, RP 11a; for ^{135m}Xe the decay branch from ^{135}I should be used
^{135}Xe	$\gtrsim$.99	~1	2.4	2.6	
^{137}Xe	.90	1.1	2.3	2.6	^{137}Xe fc: e .90±.01 w
^{138}Xe	.83	1.2	2.7	~3	^{138}Xe fc: e .83±.01
^{139}Xe	.48	2.1	3.1	~4	^{139}Xe fc: e .48±.01
^{140}Xe	.23	4.5	1.9	~5	^{140}La fi: e .0038±.0001 [5] / ^{140}Ba deduced fc: .9962±.0001; fi: e .27±.04 / ^{140}Cs deduced fc: .73±.04 / ^{140}Xe fc: e .23±.01
^{140}Cs	.73	5.5	1.9	~6	
^{140}Ba	.996	<0.1	1.9	1.9	
^{141}Xe	.051	6	7.6	10	^{141}Xe fi: e .051±.003; G: fi ≈ fc

3.2. Table A2-IV: fractional yields from ^{233}U thermal fission

Mass 87: fc for ^{87}Kr is obtained from a plot on probability paper through the experimental points. The uncertainty extrapolated from the error margins of experiments (worst case) ranges from + 7% to - 10%. Compared to a ^{89}Kr fc of $0.86 \pm .01$, the value of 0.88 obtained from the plot for fc of ^{87}Kr appears to be very unlikely. The experimental points as well as $\sigma = 0.93$ disagree with calculated data [2]. Therefore an uncertainty ranging from + 14% to - 20% should be a safe limit for fc of ^{87}Kr.

Mass 88: Only one measurement of $^{88}Br/fc$ is available. Using $\sigma = 0.64$ (mass 88 in ^{235}U thermal fission), the value of $Kr/fc = 0.94$ is obtained. With $\sigma = 0.93$ (mass 87 in ^{233}U thermal fission) Kr/fc becomes 0.83. We adopt $^{88}Kr/fc \gtrsim 0.80 \pm 20\%$, where an uncertainty of - 20% is very unlikely compared to $^{89}Kr/fc$.

Mass 90: $^{90}Kr/fc$ is deduced from the experimental data shown; these are completely consistent with G ($\sigma = 0.60$, $Zp = 36.23$), but in disagreement with the calculation of Crouch [2] ($\sigma = 0.60$, $Zp = 36.47$). The uncertainty derived from experiments is < 1%. However, since Kr/fc was not measured and fluctuations of fc around G due to the odd-even effect are possible (Amiel [3]), we adopt an uncertainty of +2 to -3 %.

Mass 133: Only measurements of I/fi exist for this mass chain. Assuming an average σ of 0.5-0.7, $I/fi = 0.15$ ($\pm$ 0.01 wa) leads to $Xe/fi \lesssim 0.01$. Even with an abnormal σ of 0.9, $Xe/fi \approx 0.05$. Therefore Xe/fc and its uncertainty should be well within the limits given in Table A2-IV. The calculated values of Crouch [2] shown in this table were derived using an earlier result of $I/fi = 0.21$ quoted by Denschlag [4] which was revised later to 0.155 (see RP 11a).

3.3. Table A2-V: fractional yields from ^{239}Pu thermal fission

Mass 87: The Gaussian parameters $\sigma = 0.72$ and $Zp = 34.80$ are obtained from a plot of the two experimental data shown in the table, and Kr/fc is deduced to be 0.991. Other possibilities of plotting G within the error limits of the measurements yield a lower limit of 0.95.

Table A2-V: Details on status of cumulative yields from ^{239}Pu thermal fission

FP	fc	uncertainty(%) for fc	chy	cy	comments and data: underlined data used to obtain fc e ... experimental, c ... calculated, w ... weighted average
^{85m}Kr	$\gtrsim$.98	$\lesssim$2*	2.3	~3	for ^{85m}Kr and ^{85}Kr see text
^{87}Kr	$\gtrsim$.98*	~3*	2	3.6	^{87}Kr fc: c .992/^{87}Br fc: e .83±.07, c .77 / ^{87}Se fc: e .33±.06, c .18 / G (see text)
^{88}Kr	~.95*	<10*	1.5	<10	^{88}Kr fc: c .95/^{88}Br fc: e .61±.03, c .49/G, pessimistic uncertainty (see text)
^{89}Kr	.86	1	2.3	2.6	^{89}Kr fc: e .86±.01
^{90}Kr	.64	2	1.4	2.4	^{90}Rb fc: c .993/^{90}Kr fc: e .64±.01, c .64/
^{90}Rb	.98*	4*	1.4	4	^{90}Br fc: e .10±.04, c .039/G (see text)
^{91}Kr	.31	3	2	3.6	^{91}Kr fc: e .31±.01
^{133}I	$\gtrsim$.98*	$\lesssim$4*	2.7	~3	^{133}I fc: c .999; fi: e .15±.03, c .17/G (see text)
^{135}I	.85	1	3.4	3.5	as evaluated by Walker, RP 11a;
^{135}Xe	>.99	<1	3.4	3.5	for ^{135m}Xe use decay branch from ^{135}I
^{137}Xe	.92	1	2.7	~3	^{137}Xe fc: e .92±.01
^{138}Xe	.85	1.2	6.5	6.6	^{138}Xe fc: e .85±.01
^{139}Xe	.55	2	5.2	5.6	^{139}Xe fc: e .55±.01
^{140}Xe	.30	2.7	1.6	3.6	^{140}Ba fc: c .997/^{140}Cs fc: c .87/
^{140}Cs	$\gtrsim$.70*	~30*	1.6	~30	^{140}Xe fc: e .30±.01, c .30/G, see text for expected fc of ^{140}Cs and ^{140}Ba
^{140}Ba	>.90*	$\gtrsim$10*	1.6	$\gtrsim$10	
^{141}Xe	.079	5	2.3	6	^{141}Xe fc: e .079±.004

Mass 88: As in the case of ^{233}U, only one measurement exists and the arguments for the uncertainty of Kr/fc shown in Table A2-V are similar. Values of 0.95-0.98 for Kr/fc are obtained from the plot.

Mass 90: The plot through the experimental data yields values for $\sigma = 0.65$, $Zp = 36.21$ and $Kr/fc = 0.98$. The uncertainty takes account of experimental errors and possible fluctuations due to the odd-even effect [3].

Mass 133: The same comments as for ^{233}U apply here.

Mass 140: The only measurement available for this mass chain is that for Kr/fc. Using $\sigma = 0.65$ from ^{235}U fission for $A = 140$, the following values are obtained:

$$Cs/fc = 0.77, \quad Ba/fc = 0.976$$

In ^{233}U thermal fission $Cs/fc = 0.73$ and $Ba/fc = 0.996$ (Table A2-IV). The lower limits and uncertainties for Cs/fc and particularly for Ba/fc are rather pessimistic, but the actual data should be reliably within these limits.

4. Detailed evaluation of some fractional yields from ^{235}U thermal fission

Tables A2-VI to A2-X show experimental data, deduced values, weighted averages (wa), values obtained from the plot and adopted values for mass chains 87, 88, 91, 139 and 140. Deduced values were obtained using wa of exp. only. Values in the column "plot" were found by drawing a best straight line by eye through experimental and and deduced fc data (generally fitted to the wa) plotted on probability paper. Values of σ and Zp were determined from this line. Generally, the experimental wa are adopted, as Amiel and Feldstein [3] have observed systematic deviations of experimental fi from a Gaussian distribution, which they ascribe to an odd-even effect.

The data of Crouch [2] and Amiel and Feldstein [3] are shown for comparison. For calculating fi assuming a Gaussian charge dispersion, Crouch [2] used values of Zp and σ deduced from an eye fit to experimental fc data plotted on probability paper. Amiel and Feldstein [3] have calculated "normal" fi from ^{235}U thermal fission using Zp and σ of Wahl et al [5] and compared them to fi evaluated from experimental data.

Table A2-VI: Fractional yields of mass 87 chain in ^{235}U thermal fission

Element (Z)	data	exp [1]	deduced values and wa: method	deduced values and wa: value	deduced values and wa: wa	a) plot	adopted	Crouch [2]	Amiel [3] from: normal	Amiel [3] from: exp
Kr (36)	fc		Se/fc+Br/fi+Kr/fi	.91±.10		.996	$.99^{+.01}_{-.02}$ a)	.998	.999	1.00
			Br/fc+Kr/fi	≤ 1.00						
	fi	.14±.01	exp	.14±.01	.14±.01	.136	.14±.01	.084	.111	.152±.013
			1-Se/fc-Br/fi b)	.23±.10						
Br (35)	fc	(1.06)	Se/fc+Br/fi	.77±.10	$.85^{+.02}_{-.04}$	.86	.85±.04 c)	.914	.888	.85
			1-Kr/fi b)	$.86^{+.01}_{-.04}$						
	fi	.47±.12	wa of exp	.45±.08	.50±.05	.53	c)	.474	.604	.35±.06
		.43±.11	1-Kr/fi-Se/fc b)	$.54^{+.05}_{-.07}$						
Se (34)	fc	.46±.06	wa of exp	.32±.05	.35±.04	.33	c)	.440	.284	.50
		.41±.07	1-Kr/fi-Br/fi b)	$.41^{+.08}_{-.09}$						
		.26±.05								
		.25±.05								
	fi		Se/fc-As/fi	.30±.05	.33±.04	.31		.392	.275	.49±.07
			1-Kr/fi-Br/fi-As/fi b)	$.39^{+.08}_{-.09}$						
As (33)	fc	(.04±.02)	exp adopted	.018±.009	.018±.009	.023		.048	.009	.01
		.018±.009								
	Zp					34.82		34.60	34.82	
	σ					.64		.66	.56	

a) for other plot with discussion of uncertainty of Kr/fc see text;

b) upper limit, assuming Rb/fi = $0^{+.04}_{-0}$;

c) note: ^{87}Br (to 2-3% per decay) and ^{88}Br (to 4-6% per decay, ie. 4-6% of ^{87}Br cumulative yield) are delayed neutron emitters (values: see S. Amiel, RP 13)

4.1. Table A2-VI mass 87 chain

Experimental data for Se/fc are discrepant and their weighted average is not reliable. Because of the large uncertainty of Br/fi, experimental data and deduced values just agree within the error limits. The values themselves are, however, not consistent with G, as shown by the deduced value of Kr/fc. The plot is obtained from a fit to wa. The adopted values are based on Kr/fi and G, from which Rb/fi $\leq$ 0.01 and Rb/fc = 1.00 are deduced.

In order to obtain a lower limit for Kr/fc, another line was drawn which is consistent with Kr/fi and the upper limit for Se/fc, but completely ignores As/fc. The following data were obtained:

Kr/fc	Kr/fi	Br/fc	Br/fi	Se/fc	Se/fi	As/fc	Zp	σ
.97	.145	.825	.34	.485	.33	.155	34.55	1.03

Although this curve with σ = 1.03 is very unlikely and disagrees with most experimental data, Kr/fc is still 0.97. Therefore the value of Kr/fc = $0.99^{+.01}_{-.02}$ should be reliable.

4.2. Table A2-VI: mass 88 chain

The situation here is very similar to that of mass 87. The plot is essentially based on Kr/fi (Kr/fc deduced) and Se/fc. The adopted values are deduced from experimental data for Kr/fi and Se/fc, and on Kr/fc obtained from the plot.

4.3. Table A2-VIII: mass 91 chain

Experimental data and deduced values are consistent and in excellent agreement with G (plot). On the other hand, there is severe disagreement with the calculated data of Crouch [2]. In his calculations, Crouch used a wrong experimental value for Sr/fi (0.30 $\pm$ 0.03 instead of 0.03 $\pm$ 0.03 [5]) and measurements of the ^{91}Sr absolute yield for Sr/fc. All experimental ^{91}Sr yield data are lower than the ^{91}Zr yield (91 chain yield) adopted by Crouch [6], but were measured relative to another chain yield and not as fractional yields. While the deviation of the measured ^{91}Sr cumulative yields from the total chain yield should be further investigated, these data are not used here.

Table A 2 - VII: Fractional yields of mass 88 chain in ^{235}U thermal fission

Element (Z)	data	exp [1]	deduced values and wa			plot[a]	adopted	Crouch [2]	Amiel [3] from	
			method	value	wa				normal	exp
Rb (37)	fc					1.00	1.00	.963	1.00	
	fi					.038		.132	.013	.01
Kr (36)	fc	1.04 ± .06	exp	$1.00^{+0}_{-.02}$	$1.00^{+0}_{-.02}$	.962	.96 ± .04	.83	.987	.99 ± .04
			Se/fc + Br/fi + Kr/fi	$1.00^{+0}_{-.18}$						
	fi	.37 ± .03	exp	.37 ± .03	.37 ± .03	.35	.37 ± .04	.28	.315	.37 ± .03
			1 - Br/fi - sc/fc	.32 ± .23						
Br (35)	fc		Sc/fc + Br/fi	.68 ± .23	.63 ± .05	.61	.59 ± .06 c)	.55	.67	.62
			1 - Kr/fi b)	$.63^{+.03}_{-.05}$						
	fi	.56 ± .23	1 - Kr/fi - Se/fc b)	.51 ± .05	.51 ± .05	.49	.47 ± .07 c)	.31	.58	.51 ± .03
			exp	.56 ± .23						
Se (34)	fc	.24 ± .06	wa of exp	.12 ± .02	.12 ± .02	.12	.12 ± .02 c)	.24	.090	.11 ± .01
		.11 ± .01	1-Kr/fi - Br/fi	.07 ± .23						
	fi	.13 ± .02				.12		.18	.089	.11 ± .01
	Zp					35.30		35.35	35.25	35.32
	σ					.68		1.20	.56	.71

a) for other possibility and discussion see text

b) upper limit, assuming Rb/fi = $0^{+.04}_{-0}$

c) note: ^{88}Br (to 5 - 6% per decay) and ^{89}Br (to 6 - 13% per decay, i.e. 4 - 8% of ^{88}Br cumulative yield) are delayed neutron emitters (values: see S. Amiel, RP 13).

Table A2-VIII: Fractional yields of mass 91 chain in ^{235}U thermal fission

Element (Z)	data	exp [1]	deduced values and wa			plot	adopted	Crouch [2]	Amiel [3] from	
			method	value	wa				normal	exp
Sr (38)	fc		Rb/fc+Sr/fi	$.96^{+.04}_{-.06}$	$1.00^{+0}_{-.02}$	1.00	1.00	.985	1.00	1.00
			Kr/fc+Rb/fi+Sr/fi	$1.00^{+0}_{-.02}$						
			Br/fi+Kr/fi+Rb/fi+Sr/fi	$1.00^{+0}_{-.02}$						
	fi	.03±.03	exp	.03±.03		.02		.18	.032	.03±.03
Rb (37)	fc	.93±.05	exp	.93±.05	.98±.02	.980	.98±.02	.804	.968	.97±.03
			1-Sr/fi	.97±.03						
			Kr/fc+Rb/fi	$.99^{+.01}_{-.02}$						
	fi	.40±.02	wa of exp	.40±.02	.39±.02	.39	.39±.02	.48	.440	.38±.03
		.39±.03	Rb/fc-Kr/fc	.34±.05						
			1-Sr/fi-Kr/fc	.38±.03						
Kr (36)	fc	.59±.01	exp	.59±.01	.59±.01	.59	.59±.01	.323	.528	.59±.01
			Rb/fc-Rb/fi	.53±.06						
			1-Sr/fi-Rb/fi	.57±.04						
			Br/fi+Kr/fi	.62±.07						
	fi	.54±.02	exp	.54±.02	.54±.02	.536	.54±.02	.285	.485	.546±.015
			Kr/fc-Br/fi	.52±.07						
			1-Sr/fi-Rb/fi-Br/fi	.50±.08						
Br (35)	fc		Br/fi≈Br/fc	.075±.07	.05±.02	.054	.05±.02	.038	.043	.044±.018
			Kr/fc-Kr/fi	.05±.02						
			1-Sr/fi-Rb/fi-Kr/fi	$.03^{+.04}_{-.03}$						
	fi	.075±.07	exp	.075±.07		.054		.037	.043	.044±.018
	Zp					36.37		36.85	36.46	36.37
	σ					.55		.76	.56	.565

Table A 2 - IX: Fractional yields of mass 139 chain in ^{235}U thermal fission

Element (Z)	data	exp [1]	deduced values and wa			plot	adopted	Crouch [2]	Amiel [3] from	
			method	value	wa				normal	exp
Ba (56)	fc		Xe/fc + Cs/fi + Ba/f1:	1.00	1.00	1.00	1.00	1.00	1.00	1.00
	fi	.011 ± .004	exp	.011 ± .004		.0065		.008	.010	.011 ± .04
Cs (55)	fc		1 - Ba/fi	.989 ± .004	.989 ± .004	.9945	.99 ± .01	.992	.990	.99
			Xe/fc + Cs/fi: 1.03±.03	1.00 $\pm^{0}_{.03}$						
	fi	.24 ± .02	wa of exp	.21 ± .02	.188 ± .014	.225	.19 ± .03	.237	.280	.206 ± .030
		.20 ± .03	1 - Ba/fi - Xe/fc	.17 ± .02						
		.18 ± .02	1 - Ba/fi - Xe/fi -I/fi	.11 ± .08						
Xe	fc	.82 ± .02	exp	.82 ± .02	.802 ± .014	.77	.80 ± .03	.755	.710	.78
			1 - Ba/fi - Cs/fi	.78 ± .02						
			I/fi + Xe/fi	.88 ± .08						
	f1	.79 ± .07	exp	.79 ± .07	.72 ± .03	.63	a)	.604	.601	.68 ± .10
			1 - Ba/f1 - Cs/fi -I/fi	.69 ± .04						
			Xe/fc - I/fi	.73 ± .04						
I	fc		I/fi ≈ I/fc	.09 ± .03	.08 ± .03	.143	a)	.151	.109	.10
			Xe/fc - Xe/fi	.03 $\pm^{.07}_{.03}$						
	f1	.12 ± .05	wa of exp	.09 ± .03		.141		.148	.108	.09 ± .10
		.07 ± .04								
	Zp					54.09		54.10	54.19	54.12
	σ					.55		.58	.56	.54

a) note: not evaluated; ^{139}I (to 6 - 14% per decay) and ^{140}I (to 14 - 50% per decay, i.e. 3 - 10% of ^{139}I cumulative yield) are delayed neutron emitters (values: see S. Amiel, RP 13)

Table A2-X: Fractional yields of mass 140 chain in ^{235}U thermal fission

Element (Z)	data	exp [1]	deduced values and wa: method	value	wa	a) plot	adopted	Crouch [2]	Amiel [3] from: normal	exp
Ba (56)	fc		Cs/fc+Ba/fi	$.976^{+.024}_{-.043}$	.97±.03	.9995	$1.00^{+0}_{+.01}$	.9956	.9997	1.00
			Xe/fc+Cs/fi+Ba/f1	$.966^{+.036}_{-.037}$						
			I/fi+Xe/fi+Cs/fi+Ba/fi	(.86±.08)						
	fi	.046±.030	exp	.046±.030	.070±.015	.039	.07±.02	.076	.052	.046±.03
			1-Cs/fc b)	.07±.03						
			1-Xe/fc-Cs/fi b)	.08±.02						
Cs (55)	fc	.93±.03	exp	.93±.03	.93±.02	.961	.93±.02	.920	.948	.95
			1-Ba/f1 b)	.954±.030						
			Xe/fc+Cs/f1	.92±.02						
	fi	.31±.02	wa of exp	.32±.02	.33±.02	.37	.33±.02	.347	.512	.33±.03
		.33±.03	1-Ba/fi-Xe/fc b)	.354±.032						
Xe (54)	fc	.60±.01	exp	.60±.01	.60±.01	.59	.60±.02	.573	.436	.62
			1-Ba/f1-Cs/fi b)	.654±.036						
			I/fi+Xe/fi c)	.494±.065						
	f1	.46±.06	exp	.46±.06	.56±.04	.49		.423	.410	.604±.040
			Xe/fc-I/f1	.566±.023						
			1-Ba/f1-Cs/f1-I/fi	.60±.04						
I (53)	fc		I/fi≈I/fc c)	.034±.021	.046±.033	.095	d)	.15	.026	.02
			Xe/fc-Xe/fi	.14±.06						
	fi	.034±.021	exp	.034±.021	.034±.021	.093		.138	.026	.02
	Zp					54.35		54.35	54.59	54.06
	σ					.65		.82	.56	.82

a) for other possibilities and discussion see text;

b) assuming La/fi≈0 (even if Cs/fc=93 is adopted, deduced La/fi<.01);

c) assuming Te/fc≈0 (assumption does not hold, if I/fc≈.15);

d) note: not evaluated; ^{140}I decays to 14-50% by delayed neutron emission (S.Amiel, RP13)

4.4. Table A2-IX: mass 139 chain

With the assumption that La/fi < 0.001 and Te/fc < 0.01 (according to G), the experimental data and the deduced values are fairly consistent, but neither agree with the plot. The deviations are in qualitative agreement with those observed by Amiel and Feldstein [3] and could be due to an odd-even effect. The values deduced from experiments are therefore adopted.

4.5. Table A2-X: mass 140 chain

Except for the values of Cs/fc and Cs/fi, experimental results and deduced values do not agree. The plotted straight line takes account of all wa data but does not agree with them. Two other plots, which ignore some of the data, were attempted: plot A was obtained from Cs and Xe data only and ignores I/fc, plot B consists of a straight line drawn through Xe/fc, which, on probability paper, passes above I/fc and Cs/fc by about equal amounts.

The results are given below:

Plot	Ba/fc	Ba/fi	Cs/fc	Cs/fi	Xe/fc	Xe/fi	I/fc	Zp	σ
A	.997	.061	.936	.336	.60	.44	.16	54.29	.80
B	1.00	.022	.978	.39	.59	.53	.06	54.37	.56

None of the plots is consistent with experimental data; the adopted values are therefore based on the experimental value of Xe/fc and the consistent data for Cs, which also have highest weight in the averages. However, a value of Ba/fc > 0.99 results from all plots, in contrast to all deduced data of Table A2-X. Also La/fi = 0.03 (deduced from wa Ba/fc = 0.97) is not consistent with G and Ba/fi = 0.046 or 0.07. Therefore the value of Ba/fc = $1.00^{+0}_{-0.01}$ is adopted.

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Appendix A3: DECAY DATA

Comparison: status - user requirements

A3.1. Abbreviations used in Appendix A3

Δ ..... accuracy, uncertainty
$T_{1/2}$.... half-life
Br..... decay branching ratio
I_γ..... gamma ray energy
E_β^{max}... end point energy of individual β's
$\bar{E}_\beta$ .... average β^- energy per decay
I_β .... β-ray intensity
Q ..... total energy released per decay
ce .... conversion electrons
I_{ce}.... absolute conversion electron intensities
α ..... total conversion coefficient
IT .... isomeric transition

A3.2. Decay data of individual FP

(i) Status and user requirements are compared in Table A3-I for individual FP decay data. Only those data are listed in this Table, for which needs have been expressed explicitly. The status of decay data is taken from individual evaluations which are indicated in the comments of Table A3-I. Unfortunately, most of the available evaluations (discussed in RP12) do not give uncertainties of recommended data. Therefore the status field had to be left blank in some cases, but references to Nuclear Data Sheets are given,where information on these data can be found.

For I_β and I_{ce} the uncertainties are given as % per decay and hence are absolute uncertainties, assuming that the uncertainty of the energy release per decay is important for users.

Absolute I_γ listed in Table A3-I is restricted to the most abundant γ-rays of a particular FP, defined as those γ-rays used for identification of a FP as well as those which are abundant enough to interfere with γ-rays of other FP in a mixed FP source. This is in

accordance with user requirements, except for life sciences: see (iv) below.

(ii) From Table A3-I the following unfulfilled requirements can be summarized. Almost all $T_{1/2}$ requirements are fulfilled, with the following exceptions:

- ^{154}Eu: not fulfilled
- ^{95}Zr, ^{125}Sb: discrepancies exceed required accuracy
- ^{89}Kr, ^{115m}In, ^{117m}Sn, ^{119m}Sn: uncertainties not evaluated

Also most of the accuracy requirements for branching ratios are met. Exceptions are (see Table A3-I for exact Br):

- ^{90}Kr and ^{91}Kr: uncertainties not evaluated
- ^{95m}Nb and ^{135m}Xe: not fulfilled

In the case of absolute I_γ many requirements are not yet met, especially those for burnup determination.

Since ce do not have large intensities per decay, all explicitly stated requirements are fulfilled, except for those cases where the presently available uncertainties are not evaluated.

For most of the β ray data no evaluated uncertainties are available. Also, $\bar{E}_\beta$ is generally not included in the references listed, while Nantel [74] does not give uncertainties.

(iii) The accuracy of gamma ray energies needed for FP identification is better than 1 keV, corresponding to a Ge(Li)-detector resolution of a few keV. This requirement is met for all FP listed in Table A3-I except for

- ^{91}Y ($\Delta E_\gamma \approx 10$ keV) and
- ^{123}Sn ($\Delta E_\gamma \approx 1$ keV)

and a few gamma rays of other FP with very low abundance. This covers all other areas requesting lower accuracy for E_γ (heat release, γ-transport).

(iv) For research work in life-sciences and agriculture information on the interaction of radiations with matter, including changes of β-spectra by absorbing material, is important. For these investigations all γ's, β's and ce's emitted by radionuclei have to be known, but high accuracy is required only for the most abundant branches. All information

on β emission, including β spectra, is needed. The types of β data included in Table A3-I should be sufficient to give information on the required and presently available accuracy.

(v) Safeguards requirements for non-destructive fuel analysis are covered by burnup requirements and are therefore not listed separately. In the case of correlations for estimating the cooling time existing FPND are more than adequate and therefore requirements are not included in Table A3-I.

(vi) I_γ of ^{140}Ba is based on the absolute value of the 537 keV γ-ray branching. The previously adopted value for I_γ(537 keV) was 24% [1]. Blachot has brought to the attention of the Panel that this value needs confirmation, as in the last evaluation of the Nuclear Data Group[25] 20% are recommended for this I_γ. The most recent (preliminary) result of Debertin [18] is 24.4 ± 0.3%, which confirms the value of [1].

(vii) In a contribution to RP 5, Tasaka and Sasamoto have compared systematically the results of different γ-emitting burnup monitors. From their study there is evidence that I_γ of the 622 keV γ ray of ^{106}Rh and the 695 keV γ-ray of ^{144}Pr could be in error by about 20%. The results of Debertin's measurements [23] should help to clarify.

A3.3 Requirements for groups of FP

(i) Review Paper 4 requests 20% accuracy on half lives of delayed neutrons groups or individual precursors (with $T_{1/2}$ > 1 sec) for failed fuel detection.

- group yields listed by S. Amiel, RP13 [4], meet these requirements.
- $T_{1/2}$ of precursors evaluated by G. Rudstam (RP12 [5], including first draft) also meet these requirements. ^{85}As, as well as the less important FP ^{84}As, $^{87,88}Se$, ^{97}Y and ^{135}Sb were not included in this evaluation.

(ii) γ-rays with E_γ of about 1-4 MeV penetrate through thick shileds, even if of low abundance. Some of the very low abundant γ-rays have still to be identified and should be included in decay studies of FP with high Q values. Most important are FP with $T_{1/2}$ of more than 100 days,

- e.g.: ^{106}Rh, ^{144}Pr (^{134}Cs, ^{154}Eu)

For Pu recycling shorter cooling times have to be considered also (see RP7). Important in this case could be e.g.:
- ^{140}La, ^{156}Eu

The accuracy required for I_γ is about 30%.

(iii) E_γ and I_γ of short lived FP are needed for high energy ($\gtrsim 2$ MeV) gammas for calculation of (γ,n) cross-sections. Accuracies required are not yet known.

(iv) For fresh fuel assay (safeguards, RP 6) the following (partially unmeasured) FP decay data are required:

$T^1/2$: 1 μs to 1 s: $T^1/2$ and I_γ within a factor 4

$T^1/2$: 1s to 1h: $T^1/2$ and I_γ to $\pm$ 10%

$$\Delta E_\gamma \approx 0.3 \text{ KeV}$$

These requirements are of low priority, since the methods concerned are not in practical use.

(v) For fission yield measurements I_γ should be known to 1% and better. Examples for FP yields measured γ-spectrometrically are: ^{85m}Kr, $^{92,93}Y$, $^{95,97}Zr$-Nb, ^{99}Mo, ^{103}Ru, ^{106}(Ru-)Rh, ^{131}I, ^{132}Te-I, ^{137}Cs, ^{140}Ba-La, ^{141}Ce, ^{143}Ce, ^{144}Ce-Pr.

(vi) For environmental considerations in cases of accidents or nuclear explosions decay data and γ-spectra are required, especially for gaseous FP, in cases where these data have not yet been measured or the FP not even been identified.

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[25] PERKER, L.K. et al, Nucl. Data Sheets 12 (1974) 343.

Table A3-I: FP decay data - status and user requirements

FP	accuracy (%) for[a]							RP	status and request for	comments[d], notes and references for status
	$T_{½}$	Br	E_β^{max}	$\bar{E}_\beta$	I_β[b]	I_γ[c]	I_{ce}[b]			
^{3}H	.1 D 10 5-10		1.1 10	1.2 10 5-10 10	0 5-10			 4 7 8	status contamination fuel handling agriculture	$T_{½}$: D within 2%; I_β = 100% status: all data [1]
^{72}Ga	1.4 5-10		 e	.07 e	 e	2-9 1	 e	 8	status life sciences	status: I_γ [10]; other data (I_γ and β data without uncertainties)[11]: ΔQ given instead of $\Delta\bar{E}_\beta$
^{82}Br	.3 D 5-10 5-10		.2 e	.03 e	.3 e	.6-2 1 1		 8 8	status industry life sciences	$T_{½}$: D within 2% status: $T_{½}$, I_γ [2a]; other data [1]
^{85m}Kr	.2 5 20	3.6 10				1.1-4 10		 4 4	status FP release fuel failure	Br: to ^{85}Kr status: Br[2], other data [8]
^{85}Kr	.6 D 5 5-10 5-10 5-10	3.6 10 5-10	.9 10 e	.8 5-10 e	.01 5-10 10 e	2 10 5-10 1 1		 4 7 8 8	status contamination fuel handling industry life sciences	$T_{½}$: D up to 4%, Br: $^{85m}Kr \rightarrow {}^{85}Kr$[f] status: Br[2], other data [1,8]
^{86}Rb	.2 5-10		.3-.7 10	.3 10	.1	.9 1		 8	status agriculture	status: $T_{½}$ [2a], I_γ [1,2a], other data [1]
^{87}Kr	.7 5-10 20	1 10 20				3.4-7 10		 4 4	status FP release fuel failure	Br: $^{87}Br \rightarrow {}^{87}Kr$[g] status: Br[4], other data [8]
^{88}Kr	.7 5-10 20	2 10 20				5-7 10		 4 4	status FP release fuel failure	Br: $^{88}Br \rightarrow {}^{88}Kr$[g] status: Br[4], other data [8]
^{89}Kr	.6 5-10 20	2 10 20				10		 4 4	status FP release fuel failure	Br: $^{89}Br \rightarrow {}^{89}Kr$[g] status: Br[4]; other data [12] (I_γ given but not ΔI_γ)

^{89}Sr	.1 5-10 5-10 5-10		<.4 10 e	.5 5-10 10 e	.01 5-10 e			 7 8 8	status fuel handling agric.,industry life sciences	status: T½ [2a], other data [1]
^{90}Kr	.6 10 10 10 10 20 20					10		 4 4 4	status FP release contamination fuel failure	Br: $^{90}Br \rightarrow ^{90}Kr \approx 85\%$[3] status: T½ [5] I_γ given in [7] without ΔI_γ
^{90}Sr	1.4 5 5-10 5-10 5-10		.4 e	.4 5-10 e	0 5-10 e			 4 7 8 8	status contamination fuel handling agric.,industry life sciences	status: all data [1]; $I_\beta = 100\%$
^{90}Y	.2 5 5-10 5-10 5-10		.13 5-10 e	.2 5-10 5-10 e	.01 5-10 e			 4 7 8 8	status contamination fuel handling agric.,industry life sciences	status: all data [1]
^{91}Kr	2.3 20 20							 4	status fuel failure	Br: $^{91}Br \rightarrow ^{91}Kr \approx 93\%$[3], $\sim \Delta Br < 20\%$ status: T½ [5]
^{91}Y	.2 5-10 5-10		.3 5	.3 5-10 5	.2 5-10 	10 5-10 		 7 8	 fuel handling industry	status: all data [1] I_γ very low, but $E_\gamma \approx 1.2$ MeV
^{95}Zr	.1 D 1-2 5 5-10 5-10 5-10		1 e	2 5-10 e	.9 5-10 e	1 1-2 10 5-10 1 1		 5 4 7 8 8	status burnup contamination fuel handling agric.,industry life sciences	T½ : D: see h); i) status: T½ [2], I_γ[2a], I_β deduced from I_γ, other data[1]
^{95m}Nb	1.1 15-28 10 5-10					20 5-10	7 5-10	 7	status fuel handling	Br: $^{95}Zr \rightarrow ^{95m}Nb$; 1); status: T½, I_{ce}, I_γ (from α)[1] Br: lower value[2a], higher value [1]

Table A3-I: continued

FP	accuracy (%) for[a]							RP	status and request for	comments[d], notes and references for status
	T½	Br	E_β^{max}	$\bar{E}_\beta$	I_β[b]	I_γ[c]	I_{ce}[b]			
^{95}Nb	.3	.2-.5	.3	.5	.04	.04			status	Br: ^{95}Zr → ^{95}Nb
	1-2					1-2		5	burnup	status: T½ [2], other data [1], Br: see ^{95m}Nb
	5					10		4	contamination	
	5-10			5-10	5-10	5-10		7	fuel handling	
	5-10					1		8	agric.,industry	
	5-10		e	e	e	1		8	life sciences	
^{99}Mo	.2 D		.3-07	.25	1.2	6-5	~1		status	T½ : D within 2%; k);l)
+^{99m}Tc	10-20			10-20	10-20	10-20	10-20	7	fuel handling	status: T½ [2a], I_γ[1,2], other data [1]
	10		e	e	e	1	e	8	life sciences	
^{103}Ru	.2			3	1.5	1.2-2	.4		status	i);k)
+^{103m}Rh	1-2					1-2		5	burnup	status: T½ [2], other data [1]
	5					10		4	contamination	
	5-10			5-10	5-10	5-10	5-10	7	fuel handling	
	10					1			agriculture	
^{106}Ru	.6		.3-.8	.3	~2	3			status	γ's from ^{106}Rh k); i)
+^{106}Rh	1-2					1-2		5	burnup	status: T½ [2], I_γ[2a], other data [1]
	5					10		4	contamination	
	5-10			5-10	5-10	5-10		7	fuel handling	
	5-10		10	10		1		8	agric.,industry	
	5-10		e	e	e	1		8	life sciences	
^{110m}Ag	.2 D			.07	2	1-2			status	T½ : D(max) 1.1%; k)
+^{110}Ag	5					10		4	contamination	status: Ag110m: T½ [2]; I_γ[2]; ΔI_β from ΔI_γ
	10-20			10-20	10-20	10 20		7	fuel handling	ΔQ (instead of $\Delta\bar{E}_\beta$) from [13]
^{111}Ag	.2		.9-1.3	.9	~3	25			status	status: all data [1]
	10-20			10-20	10-20	10-20		7	fuel handling	
	10		e	e	e	1		8	life sciences	
^{123}Sn	.3			.4	~.1	10			status	status: I_γ[2a]; ΔI_β from ΔI_γ and β to gs = 99.4%
	10-20			10-20	10-20	10-20		7	fuel handling	[14]; T½, Q ($\Delta Q \approx \Delta\bar{E}_\beta$)[14]

^{125}Sn	.3 10-20			.3 10-20	~10 10-20	~30 10-20		7	status fuel handling	status: $T_{1/2}$ [2, 14], I_γ[2a]; other data[15]: ΔQ_β given ($\approx \Delta \bar{E}_\beta$), ΔI_β from ΔI_β to gs and ΔI_γ
^{125}Sb	2 D 5 5-10			0.7 5-10	~2 5-10	1.7-5 10 5-10	2.2 5-10	 4 7	status contamination fuel handling	$T_{1/2}$: D up to 10% status: $T_{1/2}$ [2a], other data[1]
^{125m}Te	1.7 10 5-10	3 10 5-10					< 1 5-10	 4 7	status contamination fuel handling	Br: $^{125}Sb \rightarrow {}^{125m}Te$; 1) 99.7% ce[1] status: all data[1]
^{127}Sb	1.3 10-20	5/1.1 10-20		.3 10-20	$\gtrsim 2$ 10-20	1.7-10 10-20		7	status fuel handling	status: Br: $^{127}Sb \rightarrow {}^{127m}Te/{}^{127}Te$[2], other data from [16] (ΔI_γ, ΔI_β deduced from values shown; ΔQ for $\Delta \bar{E}_\beta$)
^{127m}Te	2 10-20	0.2 10-20		 10-20	.2 10-20	10-15 10-20	.2	7	status fuel handling	Br: $^{127m}Te \rightarrow {}^{127}Te$ k); status: $T_{1/2}$, Br[2], I_γ[2a] I_{ce}, I_β deduced from Br;
^{127}Te	.8 10-20			~1 10-20	 10-20	~10 10-20		7	status fuel handling	daughter of ^{127}Sb and ^{127m}Te, 1) status: I_γ[2a], other data[16] (incl. I_β, not ΔI_β)
^{129m}Te +^{129}Te	1 D 5 10-20	6.6 10 10-20		 10-20	 10-20	11-15 10 10-20	 10-20	 4 7	status contamination fuel handling	$T_{1/2}$: D 3%; Br: $^{129}Sb \rightarrow {}^{129m}Te$; ^{129}Te in equilibrium k) status: $T_{1/2}$, Br, I_γ[2a], other data[17] (no uncertainties given, but ΔI_β, $\Delta I_{ce} > \Delta$Br)
^{129}I	2.5 5-10		2.6 e	 5-10 e	0 5-10 e	10 5-10 5-10	1 5-10 e	 7 8	status fuel handling life sciences	status: $T_{1/2}$ [6], other data[17], but no uncertainties given; E_β^{max} from Q_β; I_{ce}, I_γ estimated from α given in [17]; $I_\beta = 100\%$
^{131}I	.1 D 5 5-10 5-10 5-10		.1-.3 e	.1 5-10 e	.7 5-10 e	.6-3 10 5-10 1 1	.5 5-10 e	 4 7 8 8	status FP release fuel handling agric., industry life sciences	$T_{1/2}$: D within 0.4%; includes ce from ^{131m}Xe status: $T_{1/2}$ [2a]; other data[1]
^{132}Te	1.3/1 10-20 10-20 10-20		1-2.7 e	1.4 10-20 e	1 10-20 e	.1-6 10-20 1 1	8 10-20 e	 7 8 8	status fuel handling industry life sciences	$T_{1/2}$: $^{132}Te/^{132}I$, equilibrium after 1 day k) status: $T_{1/2}$ [1,2], I_γ[2α], other data[1] I_β: $\beta^-(^{132}Te) = 100\%$, $I_\beta(^{132}Te) = 0$

Table A3-I: continued

FP	Accuracy (%) for [a] $T_{1/2}$	Br	E_β^{max}	$\bar{E}_\beta$	I_β [b]	I_γ [c]	I_{Ce} [b]	RP	status and request for	Comments [d], notes and references for status
^{133}I	.5								status	Status: all data [19], but no uncertainties given for requested data (except $T_{1/2}$);
	5					10		4	FP release	
	5-10		e	e	e	1	e	8	life sciences	
^{133}Xe	.2	.1	~1	.9	.2	1.1	~2		status	Br: $^{133}I \rightarrow {}^{133}Xe$ = 97.2%; ^{133m}Xe: 100% IT
	5	10				10		4	FP release	Status: $T_{1/2}$ [2,6], Br[2a], other data [1]
	20							4	fuel failure	
	10-20	10-20		10-20	10-20	10-20	10-20	7	fuel handling	
	10-20		e	e	e	1	e	8	life sciences	
^{134}Cs	.2				~1	.2-3			status	status: $T_{1/2}$, I_γ [2a]; ΔI_β deduced from ΔI_γ and level scheme
	1-2					1-2		5	correlation	
	5					10		4	contamination	
	5-10			5-10	5-10	5-10	5-10	7	fuel handling	
	5-10					1		8	agric., industry	
^{135}I	<.1								status	status: $T_{1/2}$ [2,9]
	5							3	kinetics	
	5							4	FP release	
^{135m}Xe	.2	13				.6			status	Br: $^{135}I \rightarrow {}^{135m}Xe \cong 9.5\%$; 1
	5	10				10		4	FP release	status: $T_{1/2}$ [9], Br [8,9], I_γ [8]
	20							4	fuel failure	
^{135}Xe	.1	1.3	1-1.7	1	2-7	.6/6	~8		status	Br: $^{135}I \rightarrow {}^{135}Xe$; ^{135m}Xe: 100% IT
	5							3	kinetics	status: $T_{1/2}$ [2,9], Br[8,9] other data [8]
	5					10		4	FP release	
	20							4	fuel failure	
	5-10		e	e	e	1	e	8	life sciences	
^{136}Cs	.2					5-10			status	ΔI_γ : except 818 keV (I_γ = 100%, ΔI_γ = 0)
	5					10		4	contamination	status: [2,2a]

^{137}Xe	.3 5 20					≥19 10			status 4 FP release 4 fuel failure	status: [8]
$^{137}Cs+$ ^{137m}Ba	.3D 1-2 5 5-10 5-10 5-10		≤.2 e	.2 5-10 e	.4/6 5-10 e	.5 1-2 10 5-10 1 1	<.1 5-10 e		status 5 burnup 4 contamination 7 fuel handling 8 agric.,industry 8 life sciences	$T_{1/2}$: D > 3%; γ's from ^{137m}Ba; i status: $T_{1/2}$ [2a], other data [8]
^{138}Xe	.5 20								status 4 fuel failure	status: [8]
^{139}Xe	1.2 20								status 4 fuel failure	status: [5]
^{140}Xe	1.2 10 20								status 4 contamination 4 fuel failure	status: [5]
^{140}Cs	.6 20								status 4 contamination	status [5]
^{140}Ba	<.1 1-2 5 5-10 5-10			~2 5-10	3.5 5-10	1.2D 1-2 10 5-10 1	3.3 5-10		status 5 burnup 4 contamination 7 fuel handling 8 agriculture	I_γ : D: see text; status: $T_{1/2}$ [2]; I_γ [18] (see text) other data [1]
^{140}La	~.1 2-3 5 5-10 5-10			.4 5-10	~3 5-10	.3-5 1-2 10 5-10 1	<.1 5-10		status 5 burnup 4 contamination 7 fuel handling 8 agric.,industry	i) status: I_γ [2a], other data [1]

Table A3-I: continued

FP	Accuracy (%) for [a] $T_{1/2}$	Br	E_β^{max}	$\bar{E}_\beta$	I_β [b]	I_γ [c]	I_{Ce} [b]	RP	status and request for	Comments [d], notes and references for status
^{141}Xe	.6								status	status: [5]
	20							4	fuel failure	
^{141}Ce	.3		.7		1.6	2	.5		status	i); status: $T_{1/2}$ [2a], other data [1] (uncertainty of $\bar{E}_\beta$ not evaluated $\rightarrow \Delta E_\beta^{max}$ given in table).
	1-2					1-2		5	correlation	
	5-10			5-10	5-10	5-10	5-10	7	fuel handling	
^{143}Pr	.2				.2				status	status: $T_{1/2}$ [2a], other data [20]
	5-10			5-10	5-10			7	fuel handling	
$^{144}Ce+$	.2		.1-.8	.5	~1	3-8	.3		status	k); i)
^{144}Pr	1-2					1-2		5	burnup	status: $T_{1/2}$ [2], I_γ [2a], other data [1]
	5-10			5-10	5-10	5-10	5-10	7	fuel handling	
	5-10		10	10		1		8	agric. ,industry	
	5-10		e	e	e	1	e	8	life sciences	
^{147}Nd	.3				≥10	8-10			status	status: $T_{1/2}$; I_γ [2a], other data [21] without uncertainties; ΔI_β estimated from ΔI_γ and level scheme of [21]
	5-10			10-20	10-20	10-20		7	fuel handling	
^{147}Pm	.1		.3	.3	0				status	status: [1]; I_β = 99.992 %
	5-10			5-10	5-10			7	fuel handling	
	5-10		5-10					8	industry	
^{149}Pm	.1					6			status	status: $T_{1/2}$, I_γ [2a]
	10-20			10-20	10-20	10-20		7	fuel handling	
^{151}Sm	5								status	status: $T_{1/2}$ [2a]
	5-10			10-20	10-20		5-10	7	fuel handling	

^{153}Sm	.4D 10-20 5-10		.5-1	10-20	5 10-20	2-5 10-20 1	5 10-20	7 8	status fuel handling agriculture	T ½ : D within 2% status: T ½ [2a], I_γ [2a, 22], other data [22] (but no uncertainties); ΔI_β, ΔI_{ce} deduced from [22] and ΔI_γ
^{154}Eu	6D 1-2 5-10			5-10	5-10	1-3 1-2 5-10		 5 7	status correlation fuel handling	T ½ : D e.g. 8.5 (adopted) and 16 years status: T ½ [2], I_γ [2a]
^{155}Sm	5-10		e	e	e	1	e	 8	status life sciences	
^{155}Eu	.2 5-10 5-10 5-10		 e	5-10 e	5-10 e	3-4 5-10 1 1	5-10 e	 7 8 8	status fuel handling industry life sciences	T ½ : 1.8 years obviously wrong status: [2a]
^{156}Eu	.2 10-20			10-20	10-20	10-12 10-20		 7	status fuel handling	status: [2a]

a) Status: D ... some discrepancies among experimental data exist!
blank, if reeds are given: not included in evaluations which give uncertainties (except Nuclear Data Sheets up to 1965).

b) Only most abundant β and ce considered in status; uncertainty given in % per decay, which is equivalent to absolute uncertainty.

c) Range of accuracy for most abundant γ's given.

d) D ... discrepancies
Δ ... uncertainty

e) For research work the accuracy should be as high as possible. All information on β decay is requested (see text).

f) Assuming 100% branching $^{85}Br \rightarrow {}^{85m}Kr$. If available, cumulative yields for ^{85}Kr should preferably be used.

g) The precursor is delayed neutron emitter. Its delayed neutron branch to ^{A-1}Kr has to be considered also. The use of cumulative yields could be preferable, but ultimately β^- and delayed neutron branches will be used in inventory calculations together with independent yields.

Table A 3 - I: continued

h) A discrepancy of 2.3% exists between 2 measurements; see: K. Debertin et al. [23] (these Panel proceedings, Vol. 3)

i) Measurements of γ-ray emission probabilities (absolute intensities) to 1% accuracy at the Physikalisch-Technische Bundesanstalt, Braunschweig, FRG, are completed or in progress; see: K. Debertin et al. [23].

k) Daughter in equilibrium with parent at any time of interest; $T_{1/2}$ of daughter unimportant; radiation intensity per decay of parent is essential information and given in the table (Br is ignored).

l) Daughter reaches equilibrium with parent after about 10 $T_{1/2}$ of daughter in an initially pure source of the parent nuclide; at the end of long irradiations equilibrium is generally reached.

Appendix A4: NEUTRON REACTION CROSS-SECTIONS

Comparison: status - user requirements

A4.1 Abbreviations

A number of abbreviations are used, particularly in the tables, which are explained below:

σ .... neutron capture cross-section(s), specifically:
- σ_{th} ... in the thermal energy range
- σ_o ... at 2200 m/sec (0.0253 eV)
- $g\sigma_o$ ... averaged over a thermal maxwellian spectrum
- σ(E)... as function of incident neutron energy
- σ(fast) averaged over a fast reactor spectrum.

RI ... infinite dilution resonance integral for neutron capture
res .. resonance(s)
respars.. resonance parameters
E .... energy (of incident neutrons or resonances)
exp .. experimental

A4.2 Neutron capture cross-sections for thermal reactors

(i) The data of interest for thermal reactors are represented by σ_{th} and RI. User needs and data status are compared in Table A4-I. It can be seen that several requirements for σ_{th},RI are not fulfilled.

(ii) User requirements are expressed for integral σ-data such as σ_{th} and RI or pile -σ. However, a knowledge of σ(E) is required to an accuracy sufficient to allow the calculation of an average σ for any thermal reactor spectrum ranging from D_2O moderated reactors to HTGR. On the other hand, integral data may be sufficient, if they can be used for all thermal reactor spectra within the requested accuracy limits:

RP3: σ(E) is required in the thermal range (indicated in column 11 of Table A4-I), particularly for ^{135}Xe and ^{149}Sm.

RP5: σ_o or $g\sigma_o$ is sufficient for the thermal range. For the epithermal range σ(E) is required, if RI cannot be used within the requested accuracy for different reactor spectra (where $\phi(E)$ deviates from 1/E).

Table A4 - I: Neutron capture cross-sections for thermal reactors: status and user requirements

FP	type of σ	accuracy required for[a] RP3 (%)	RP4 (%)	RP5,6 (%)	RP5,6 (b)	RP7 (%)	data status[b] accuracy of σ_c (%)	accuracy of σ_c (b)	E-range of resolved res	E-range for requirements[c]	Comments
^{99}Tc	σ_{th}	20					10	2		< 1 eV	
	RI	15					10		5.6-280eV	1-500 eV	79 res known
^{103}Rh	σ_{th}	6					4			< 10 eV	
	RI	50					5		1eV-4.1keV	1-200 eV	275 res known
^{109}Ag d)	σ_{th}		} 20				15	0.6		< 1 eV	
	RI						13	7	5eV-2.5keV	1-300eV	81 res known
^{110m}Ag	σ_{th}		} 20				} 14	11			only one exp on pile -σ
	RI										
^{133}Xe	σ_{th}				} 150		} 50	90			only pile -σ available
	RI										
^{133m}Xe	σ_{th}				} 10^4						no data available
	RI										
^{133}Cs	σ_{th}	15	} 20	2		} 5-10	5	1.5		< 1eV	
	RI	10		2			7	30	5.9eV-3.5keV	1-500 eV	164 res known
^{134}Cs	σ_{th}		} 20	3		} 5-10	9	12			only pile -σ available
	RI			25			factor 20				
^{135}Xe	σ_{th}	8	5				3			< 4eV	no significant res except .084 eV
	RI	100					7		.084eV		

^{135}Cs	σ_{th} RI		} 20			6 20[e]	0.5 10[e]			RI: disagreements[e] no respars available
^{141}Pr	σ_{th} RI			} 2.8		2.6 1.4[e]	0.3 0.2[e]	85eV-10keV	< 20eV 20eV-2keV	RI: evaluations disagree[e] 120 res known
^{143}Pr	σ_{th} RI			} 30			10 25			
^{143}Nd	σ_{th} RI	6 30		2 6		3 20	10 30	55eV-5.5keV	< 10eV 10 - 500eV	negative res at- 6eV ~100 res known
^{144}Ce	σ_{th} RI			2 6		10 12	0.1 0.3			no respars available
^{144}Nd	σ_{th} RI			2 6			0.3 0.5	0.37-19 keV	< 0.5 eV 0.5eV-10keV	35 res known
^{145}Nd	σ_{th} RI			2 6			2[e] 35	4eV-4.6keV	< 0.5eV 0.5eV-1keV	evaluations disagree[e] 191 res known
^{146}Nd	σ_{th} RI			2 6			0.1 0.5	0.36-17 keV	< 0.5 eV 0.5eV-10keV	44 res known
^{147}Nd	σ_{th} RI			} 30						only preliminary data available
^{147}Pm	σ_{th} RI	15 8			} 5-10	7 7	13 150	5.4-317eV	< 1eV 1eV-200eV	negative res at -1.8eV 41 res known
^{148}Nd	σ_{th} RI			2 6			0.2 1	95eV-12keV	< 0.5eV 0.5eV-3keV	66 res known

Table A4-1: cont'd "Neutron capture cross-sections for thermal reactors: status and user requirements"

FP	type of σ	accuracy required for[a] RP3 (%)	RP4 (%)	RP5,6 (%)	RP5,6 (b)	RP7 (%)	data status[b] accuracy of σ_0 (%)	accuracy of σ_0 (b)	E-range of resolved res	E-range for requirements[c]	Comments
^{149}Sm	σ_{th} RI	20		f	f		3	1200	0.1-249eV	≲ 20eV	thermal extended to 20eV, RI unimportant
^{150}Nd	σ_{th} RI				2 6			0.2 2	79eV-14keV	< 0.5eV 0.5eV-3keV	78 res known
^{151}Sm	σ_{th} RI	8 40		f	f		15 20		1.1-13eV	< 0.5eV 0.5 - 30eV	negative res at -0.1eV, 10 res known
^{152}Sm	σ_{th} RI	20 10		f	f		3 5	3 15	8eV-5keV	< 1 eV 1-500eV	~ 100 res known
^{153}Sm	σ_{th} RI				~10^3						no data available estimates: ~factor 10
^{153}Eu	σ_{th} RI			3 10		5-10	~20 12	80 200	0.4-100eV	≲ 1 eV 0.4-100eV	σ_{th}: discrepant data ~ 90 res known
^{154}Eu	σ_{th} RI			3 10		5-10	~25	400			only one exp on pile -σ

a) Principally σ has to be known as function of incident neutron energy, but requirements are generally expressed for an integral σ derived from σ (E). However, $g\sigma_0$, RI or pile-σ may be sufficient, if the variation with reactor neutron spectrum (including HTGR) is within the requested uncertainty. With these limitations, the requirements are (see also text):

RP3: σ(E) required for thermal range shown in column 11 of the table, particularly for ^{135}Xe and ^{149}Sm.

RP4 and RP7: pile -σ may be sufficient.

RP5: $\sigma(E)$ and res pars desirable, except for low accuracy requirements.

b) σ_{th}: generally accuracy of σ_0 or for Maxwellian averaged σ ($g\sigma_0$); note that $\sigma(E)$ may be required;
RI: accuracy of RI (above 0.5 eV) calculated from res pars and compared to integral measurements;

c) σ_{th}: energy range, for which $\sigma(E)$, if requested, has to be known;
RI: energy range of resolved resonances required for description of RI or $\sigma(E)$ (except ^{135}Xe: range given for $\sigma(E)$)

d) Cross-section for formation of 252 d ^{110m}Ag;

e) Discrepancies among experiments and/or evaluations (shown in RP10, table III) exceed the uncertainty shown.

f) σ required, but definite accuracy not yet known (see section 3.2, item (viii)).

RP4 and RP7: $g\sigma_o$ (with the possible exception of ^{135}Xe) and RI may be sufficient for the FP listed in Table A4-I. For other FP not listed but discussed in sections 3.2 and 7.2 the knowledge of pile-σ should be sufficient.

If $\sigma(E)$ is required, the accuracy needed is inversely proportional to the neutron energy.

(iii) Column 11 of Table A4-I shows the energy range for $\sigma(E)$ required by users.

The thermal range is chosen to include the part of $\sigma(E)$ which contributes significantly to thermal captures (particularly important for reactor design).

Generally group cross-section data are used in large codes. However, the Panel recommends (section 7.3) to describe in evaluations the low energy part of a cross-section curve in the epithermal range in terms of resolved resonances. Therefore the energy range shown in column 11 of Table A4-I is chosen to include about 20 resonances or is extended to the highest resonance with supposedly significant contribution.

(iv) Data needs for safeguards are generally covered by burnup requirements. Additional requirements are:

- The ratio of capture cross-section uncertainties of ^{133}Cs to ^{134}Cs should be 1:3. In order to achieve the accuracy of burnup determination required by safeguards, the captures in ^{133}Cs (^{134}Cs) have to be calcuated to 1% (3) accuracy for any thermal reactor spectrum. This requires a knowledge of $\sigma(E)$ of ^{133}Cs and ^{134}Cs to the appropriate accuracy.

- Requirements for σ to be used in correlation studies depend upon the accuracy required for the result, which has not been determined at the time of the present Panel. Sensitivity studies aiming at 1% accuracy for a particular case quoted in RP 6, resulted in the following requirements for $\sigma\gamma$ of individual FP:

^{82}Kr	20	^{130}Xe	20%
^{83}Kr	3%	^{131}Xe	1.4%
^{129}I	20%	^{135}Xe	10%

(v) The data status is explained in Table A4-I. In the thermal range the the accuracy for $\sigma(E)$ is given, if another representation is not sufficient (e.g. ^{135}Xe, ^{149}Sm).

The status of the σ-data was supplied by P. Ribon, who considered also contributions by Walker, by Pope and Story, and by Sakata and Nagayama (see RP10). Additional values (e.g. res ranges) were taken from BNL-325, 3rd edition, Vol. I (June 1973).

A4.3 Neutron capture cross-sections for fast reactor applications

(i) User requirements and data status are compared in Table A4-II for fast reactors. The table shows that the majority of requirements is not fulfilled.

(ii) For the calculation of the Na-void effect in fast reactors (RP 3), $\sigma(E)$ is required in the energy range 0.1 - 100 keV for all FP contained in Table A4-II.

(iii) The data status is given for three different, partly overlapping, energy ranges, which are explained in Table A4-II. σ_c in the range of resolved resonances (up to several keV) may contribute significantly to the total σ_c (fast). A knowledge of average resonance parameters is important for the calculation of σ_c in the 10-100 keV range.

Presently available accuracies of data in the columns headed "res range" and "fast range" were supplied by P. Ribon together with comments on available experimental data and res-pars (section A4.4). The status of $\sigma(E)$ for $E \geq 0.1$ MeV obtained from the Bologna FP cross-section library was communicated by V. Benzi.

A4.4 Comments to data status in Table A4-II

^{95}Mo: $\bar{\Gamma}_\gamma$ is known to 7%, but dependence of S, $\bar{\Gamma}_\gamma$ and $\bar{D}$ on (J, π, E) is badly known;

only one measurement of $\bar{\sigma}$ in the 10 keV range with slowing down spectrometer (SDS).

^{97}Mo: same remarks as for ^{95}Mo.

Table A4-II: Neutron capture cross-sections for fast reactor applications

status and user requirements

FP	accuracy required a)			accuracy achieved b)				notes
				res range		fast range	≥0.1MeV σ(E)	
	RP3	others			E_{max}			
	(%)	RP	accuracy	(%)	(keV)	(%)	(%)	
^{95}Mo	30				2.1	30	v.poor	c)
^{97}Mo	30				1.9	30	v.poor	c)
^{98}Mo	35			10	9	50	poor	c) d)
^{99}Tc	20			10	0.28	40	theo	d) e)
^{100}Mo	35			7	4.7	30	50	c) d)
^{101}Ru	20				0.67	40	theo	c)
^{102}Ru	25			30	1.3	30	v. poor	e)
^{103}Rh	20			5	4.1	10	30	
^{104}Ru	30			30	1.1	40	v.poor	d)e)
^{105}Pd	20				0.8	30	theo	e)
^{106}Ru	35	5	25-50 mb			fact.2	theo	
^{107}Pd	25			fact.5		fact.2	theo	e)
^{109}Ag		4	20%	13	2.5	25		
		7	10-20%					
^{131}Xe	30			5	4	40	theo	c)e)
^{133}Cs	20	4	20%	7	3.5	20	30	
		7	5-10%					f)
^{134}Cs		4	20%			fact.2		
^{135}Cs	30	4	20%	30		fact.2	theo	e)
^{137}Cs	40	5	25-50mb			fact.2		
^{139}La	40			5	10.4	<40		
^{141}Pr	35			15	10	20	30	d)
^{143}Nd	30	5	25-50 mb	20	5.5	30	theo	c)
^{144}Ce		5	25-50 mb			fact.2		

Table A4-II: (continued)

FP	accuracy required[a]			accuracy achieved[b]				notes
				res range		fast range	≥0.1MeV σ(E)	
	RP3	others			E_{max}			
	(%)	RP	accuracy	(%)	(keV)	(%)	(%)	
^{144}Nd		5	25-50 mb		19.4	35		
^{145}Nd	35	5	25-50 mb	15	4.6	30	theo	
^{146}Nd		5	25-50 mb	16	17	30		
^{147}Pm	25	7	5-10%	7	0.3	40	theo	c)d)
^{148}Nd		5	25-50 mb	5	12	~20		
^{149}Sm	30				0.25	30	theo	c)
^{151}Sm	25				<0.1	50	theo	e)
^{151}Eu		CR	10%			40	30	e)f)
^{152}Sm	55			5	5.1	<50		
^{152}Eu		CR	10%			fact.2		e)f)
^{153}Eu	40	7	5-10%		0.1	40	theo	d)f)
		CR	10%					
^{154}Eu		7	5-10%			fact.2	theo	e)f)
		CR	10%					
^{155}Eu	55					fact.2		e)

a) σ (E) required in the energy range 0.1 keV - 5 MeV; a fast reactor spectrum averaged σ is sufficient, if its variation is within the requested accuracy for different fast reactors.

CR ...requirement for control rod purposes in the energy range 10 keV - 1MeV

b) Detailed comments are given in the text

res range: range of resolved res; the accuracies are for RI above 0.1 keV, obtained from res pars;

E_{max}: energy of highest resolved res.

Table A4 - II (continued)

fast range: energy range above highest resolved resonance up to a few MeV as given by Ribon (see text).

$\geq$0.1 MeV: uncertainties given for Bologna library (see RP10)

poor ... estimated uncertainty of $\sigma(E) > 50\%$

v.poor ..very poor: theoretical estimate + few experimental data

theo ... theoretical estimate only.

c) The requested accuracy ($\gtrsim 30\%$) should be achieved (or improved to 20%) from a better knowledge of average resonance parameters in the 100 keV range and of level schemes of the target nuclei.

d) New evaluations based on available experimental data should allow a resolution of discrepancies.

e) A better knowledge of res-pars should improve the accuracy of calculated cross-sections.

f) An accuracy of ~10% can only be achieved (or improved, if required) from direct measurements.

g) σ for formation of 252 day ^{110m}Ag.

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^{98}Mo: same remarks as for ^{95}Mo ($\bar{\Gamma}_\gamma$ to 5%);
8 sets of exp data for $\bar{\sigma}$ above 1 keV.

^{99}Tc: $\bar{\Gamma}_\gamma$ known to 20%; only 4 res below 0.1 keV;
only 1 SDS measurement above 1 keV.

^{100}Mo: Probably a strong dependence of $\bar{\Gamma}_\gamma$ on parity;
7 sets of exp data above 1 keV.

^{101}Ru: $\bar{\Gamma}_\gamma$ known to 5%;
no exp data for σ_c.

^{102}Ru: no $\bar{\Gamma}_\gamma$ value;
3 sets of exp data above 1 keV.

^{103}Rh: well known res pars;
many σ_c measurements; improvement of accuracy of σ_c, if required, can only be obtained from direct measurements of σ_c.

^{104}Ru: 5 sets of exp data for σ_c.

^{105}Pd: $\bar{\Gamma}_\gamma$ known to 7%;
1 measurement of σ_c above 1 keV.

^{107}Pd: no experimental data.

^{133}Cs: res pars well known;
10 sets of exp data on σ_c above 1 keV.

^{134}Cs: no exp data on σ_c; radioactive nucleus: differential measurement impossible.

^{135}Cs: no exp data available.

^{141}Pr: more than 10 sets of exp data on σ_c above 1 keV.

^{143}Nd: $\bar{\Gamma}_\gamma$ known to 4% accuracy;
no exp data for σ_c

^{145}Nd: $\bar{\Gamma}_\gamma$ known to 6% accuracy;
no exp data for σ_c.

^{147}Pm: $\bar{\Gamma}_\gamma$ known to 8% accuracy, but highest res at 0.3 keV;
1 measurement of σ_c above 1 keV.

^{149}Sm: $\bar{\Gamma}_\gamma$ known to 4% accuracy;
1 measurement of σ_c above 1 keV.

^{151}Sm: no measurements of σ_c above 1 keV.

^{151}Eu: $\bar{\Gamma}_\gamma$ known to 6% accuracy;
6 sets of exp data above 1 keV.

^{152}Sm: $\bar{\Gamma}_\gamma$ known to 10% accuracy;
no exp data above 1 keV.

^{152}Eu: 1 measurement of σ_c above 1 keV; differential data can only be obtained from nuclear explosions.

^{153}Eu: 4 sets of exp data above 1 keV.

^{154}Eu: no measurements in fast range.

Appendix A5: FP DECAY HEAT

Since no sensitivity studies on the accuracy required for individual FPND have yet been performed, this appendix can serve as the basis for such studies. Presented are user requirements for total decay heat after reactor shutdown as well as the results of Devillers' study (appendix to RP 4) on FP that contribute significantly to the total decay heat, and the status of their half lives. Together with the status on cumulative yields and effective decay energies more detailed requirements should be worked out in future studies.

A5.1 User requirements on total decay heat

User requirements of the accuracy to which the total decay heat after reactor shutdown has to be known, is presented in Table A5-Ia as was observed by the Panel.

If we take an accuracy of $\pm$ 10% as the target for all times after shutdown up to several days, with $\pm$ 5% as a long term aim, and bear in mind that FP contribute only 40% of the total afterheat at 1 s cooling time, and 50% at about 10s, we arrive at the accuracy targets presented in Table A5-Ib for the FP contribution to the total afterheat. The values are given for the main fissile isotopes. For other fertile and fissile isotopes such as ^{238}U and ^{241}Pu the accuracies required are about a factor of 4-5 less (for individual FPND this means a difference only in fission yields).

A5.2 Contribution of individual FP to afterheat

The different cases computed by Devillers (RP 4, appendices) are summarized in graphical form in Table A5-II. Some of the FP listed in the table contribute significantly to the total heat released only in the case of ^{233}U thermal fission or ^{239}Pu fast fission (difference in mass yield distribution).

Table A.5-II together with the footnotes is essentially self-explanatory. It should give a good idea which FP have to be considered in more detailed studies.

In accordance with the Panel's conclusions the half lives of FP contributing only up to about 100 s to afterheat are required with lower accuracy than > 100 s which is indicated by (+) in Table A5-II (meaning: decay property is less important). The significance of other decay properties is indicated according to Devillers' calculations.

Precursors of important FP are indicated only in those cases where they have a significant influence on the time range the daughter contributes to the decay heat.

Current Nuclear Data Sheets were consulted in order to obtain the most recent status of half-lives. The references quoted for half-lives indicate also the latest update of other decay data by the Nuclear Data Group.

References:

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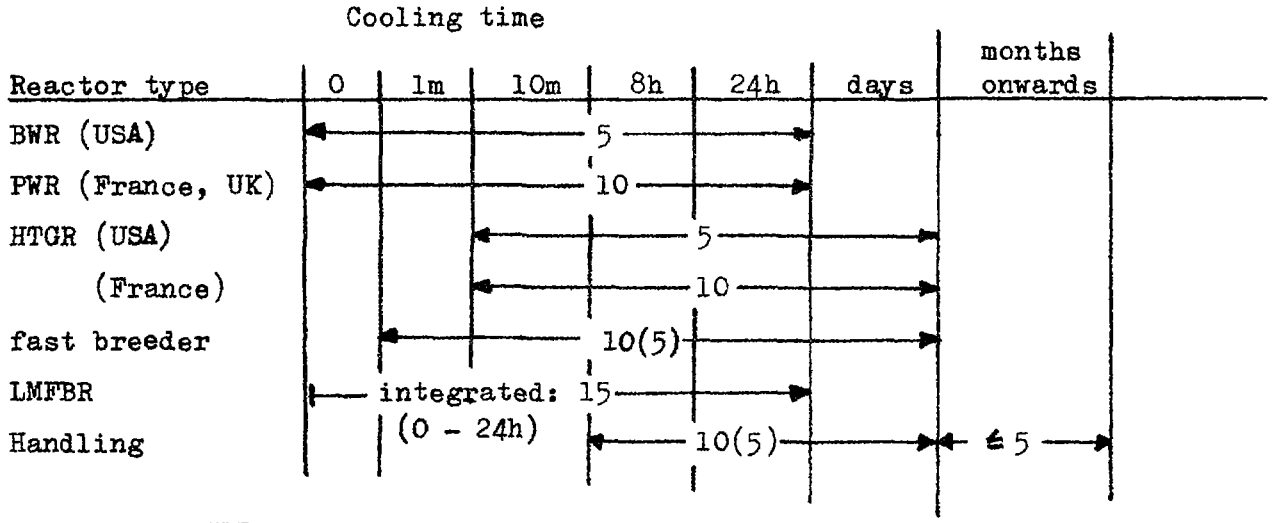
[24] MARTIN, M.J., Nucl. Data B2-4-12 (1967)
Uncertainty deduced by M. Lammer from data shown.

Table A5-I.a: Accuracy to which the total heat released after shutdown has to be known for different reactor types as a function of cooling time[a)]



a) Accuracy is given in %; higher accuracies given in brackets are a long term aim.

Table A5-I.b: Accuracy required for the energy released by fission products as a function of cooling time[a)]

	Cooling time: 1s	10s	100s ≈1.7m	10^4s ≈2.8h	10^5s ≈28h	10^6s ≈12d	10^7s ≈116d	≥10^8s ≈3.2y
thermal ^{235}U, ^{239}Pu	25(12)	20(10)	10 (5)					≤ 5
^{233}U			10 (5)					≤ 5
fast ^{235}U, ^{239}Pu	integrated: 15 (0 – 24h)		10 (5)				b)	≤ 5

a) Accuracy is given in %; higher accuracies given in brackets are a long term aim.

b) ≤ 5% accuracy required from 30 days onwards.

Table A.5 - II: FPND important for FP energy release after reactor shutdown.

FP[a]			decay property[b]					half-life				cooling time[c]; [f]									
									uncertainty			1 s	10 s	10^2 s	10^3 s	10^4 s	10^5 s	10^6 s	10^7 s	10^8 s	10^9 s
A	I	S	H	B	β	γ	E	value[c]	(%)[d]	e)	Ref			≈1.7 m	≈17 m	≈2.8 h	≈28 h	≈12 d	≈116 d	≈3.2 y	≈32 y
84	g	Br	+		+	+	+	31.7 m	0.3		7										
85	g	Kr	+		+	+		10.73 y	0.6		2,4										
86	g	Br	(+)		+	+	+	55.7 s	0.9		3										
87	g	Br	(+)	(+)	+	+	+	55.8 s	0.6		3										
	g	Kr	+		+	+		76.3 m	0.7		4										
88	g	Br	(+)	(+)	+	+	+	16.0 s	1.3		3										
	g	Kr	+		(+)	+		2.80 h	0.7		4										
	g	Rb	+		+	+		17.8 m	0.6		4										
89	g	Kr	+		+	+		3.18 m	0.6		6										
	g	Rb	+		+	+		15.2 m	0.7		6										
	g	Sr	+		+			50.52 d	0.1		2										
90	g	Kr	(+)	+	+	+	+	32.3 s	0.6		3										
	m	Rb	+	+	+	+	+	4.27 m	2		8										
	g	Rb	+		+	+		2.57 m	2.6		8										
	g	Sr	+		+			28.6 y	1.4		2										
	g	Y	(+)		+			64.06 h	0.2		5										
91	g	Rb	(+)		+	+	+	58.2 s	0.6		3										
	g	Sr	+	+	+	+		9.6 h	1.3	(D)	9										
	m	Y	+			+		49.71 m	0.1		9										
	g	Y	+		+			58.51 d	0.3		2,9										
92	g	Rb	(+)			+	+	4.50 s	0.7		3										
	g	Sr	+		(+)	+		2.71 h	0.4		10										
	g	Y	+		+	+		3.54 h	0.3		10										
93	g	Rb	(+)	(+)	+	+	+	5.81 s	0.7		3										
	g	Sr	+		+	+		7.36 m	2.0		3										
	g	Y	+		+	(+)		10.1 h	2.0	D	11										
94	g	Sr	(+)		+	+	+	75.6 s	1.2		3										
	g	Y	+		+	(+)		18.7 m	6.4	D	3										
95	g	Sr	(+)		+	+	+	28 s	11		3										
	g	Y	+		+	(+)		10.6 m	3		3										
	g	Zr	+	(+)	+	+		63.98 d	0.1	D,W	2										
	g	Nb	+		+	+		35.05 d	0.3		2										
96	g	Y	+		+	+		2.3 m	4.4		12										
97	g	Zr	+	+	+	+		17.0 h	1.2		13										
	m	Nb				+		60 s			13										
	g	Nb	+		+	+		72.1 m	1.0	h	13										
98	g	Y	(+)		+	+	+	2.3 s	?	D	1										
	g	Zr	+					30.7 s	~1	i)	14										
	g	Nb	(+)		+		+	2.8 s	7.2		14										
99	g	Mo	+		+	+		66.0 h	0.2	D	2										
100	g	Nb	(+)		+	(+)	+		?	k)											
101	g	Nb	(+)		+	(+)	+	7.0 s	4.3		16										
	g	Mo	+		+	+		14.62 m	0.4		16										
	g	Tc	+		+	+		14.2 m	0.7		16										
102	g	Mo	+					11 m	-		1										
	g	Tc			+	+		5.3 s			1										
103	g	Mo	(+)		+	+	+	60 s	-		1										
	g	Ru	+		+	+		39.35 d	0.2		2										
104	g	Tc	+		+	+		18 m	-		1										
105	g	Mo	(+)		+	+	+	42 s	7.2		17										
	g	Tc	+		+	+		7.8 m	2.6		17										
	g	Ru	+		+	+		4.44 h	1.2		17										
106	g	Tc	(+)		+	(+)	+	36 s	-		1										
	g	Ru	+					368.3 d	0.6		2										
	g	Rh			+	+		30.35 s			1										

Table A.5 - II: cont'd

FP[a]			decay property[b]					half-life				cooling time[c,f]									
								value[c]	uncertainty (%)[d]	e)	Ref	1 s	10 s	10^2 s	10^3 s	10^4 s	10^5 s	10^6 s	10^7 s	10^8 s	10^9 s
A	I	S	H	B	β	γ	E							≈1.7 m	≈17 m	≈2.8 h	≈ 28 h	≈12 d	≈116 d	≈3.2 y	≈32 y
125	g	Sb	+		(+)	+		2.75 y	1.5		2										
129	g	Sb	+		(+)	+		4.32 h	0.7		18										
131	g	Sb	+	(+)	+	+		23.0 m	-		1										
	m	Te	(+)	(+)				30 h	6.7		2										
	g	Te	+		+	+		25 m	-		1										
	g	I	+		+	+		8.04 d	0.2	D	2										
132	g	Sb	+		+	+		3.08 m	2.2		3										
	g	Te	+		+	+		78 h	2.6		2										
	g	I	+		+	+		2.285 h	0.5	D,W	2										
133	g	Sb	+	+	(+)	+		2.7 m	13		3										
	m	Te	+		(+)	+		55.4 m	0.8		19										
	g	Te	+		+	+		12.45 m	2.3		19										
	g	I	+	(+)	+	+		20.9 h	1		2										
	g	Xe	+		+	+		5.29 d	0.2		2,4										
134	g	Te	+		+	+		43 m	-		1										
	g	I	+		+	+		53.2 m	-		1										
	g	Cs	+		+	+		2.062 y	0.3	W,D	2										
135	g	Te	(+)		+	+	+	19.2 s	1		3										
	g	I	+	(+)	+	+		6.585 h	0.1	W	2										
	g	Xe	+		+	+		9.172 h	0.1	W	2										
136	?	I	(+)		+	+	+	83 s	4	g)	1,3										
137	g	I	(+)		+	+	+	24.6 s	0.5												
	g	Xe	+		+			3.83 m	0.3		4										
	g	Cs	+	+	+			30.17	0.4	D	2										
	m	Ba				+		2.552 m			4										
138	g	I	(+)		+	+	+	6.6 s	3		3										
	g	Xe	+		+	+		14.17 m	0.5		4										
	g	Cs	+		+	+		32.2 m	0.3		4										
139	g	Xe	(+)		+	(+)	+	40.4 s	1.3		3										
	g	Cs	+		+	+		9.40 m	1.3		20										
	g	Ba	+		+			84.9 m	1.3		20										
140	g	Cs	(+)		+	+	+	64.0 s	0.7		3										
	g	Ba	+		+	+		12.789 d	0.1		2										
	g	La	+		+	+		40.27 h	0.2	(D)	5,2										
141	g	Cs	(+)		+	(+)	+	25.2 s	2		3										
	g	Ba	+		+	+		18.27 m	0.4		21										
	g	La	+		+			3.93 h	1.6		21										
	g	Ce	+		+	+		32.50 d	0.2	(D)	2										
142	g	Ba	+		+	+		10.7 m	1		22										
	g	La	+		+	+		92.7 m	0.8		22										
143	g	La	+		+			14.32 m	-		1										
	g	Ce	+		+	+		33.0 h	0.6	W,D	2										
	g	Pr	+		+			13.57 d	0.2		2										
144	g	La	(+)		+	+	+	40.3 s	0.8		3										
	g	Ce	+		+	+		284.5 d	0.2		2										
	g	Pr			+	+		17.28 m			5										
145	g	Pr	+		+			5.98 h	0.4		23										
146	g	Pr	+		+	+		24.2 m	0.9		24										
147	g	Nd	+		+	+		11.01 d	0.2	(D)	2										
	g	Pm	+		+			2.6234 y	0.1		2										
154	g	Eu	+		(+)	+		8.5 y	6	D	2										
156	g	Eu	+		+	+		15.16 d	0.2		2										
% of total FP decay heat not listed												<55	<50	<43	<26	<19	<12	<8	<6	<2.3	<1.3

Table A.5 - II: cont'd.

Explanations

a) A .... mass number
I .... isomeric state: g,m = ground, metastable state
S .... element symbol

b) H .... half life
B .... branching in decay to daughter product
β .... β-decay data
γ .... γ-decay data
E .... mainly effective total energy ($E_\beta + E_\gamma$) released required
+ .... knowledge of the decay property is required
(+)... decay property is less important

c) s .... seconds
m .... minutes
h .... hours
d .... days
y .... years

d) percent error rounded upwards, i.e.: 0.1 means: ≤ 0.1%
?..... value and uncertainty questionable
blank.... half-life unimportant
- ..... uncertainty not evaluated

e) quoted uncertainty to be used with caution

W .... warning: only one measurement
D .... discrepancies among experimental values exceed uncertainty quoted in table
(D) .. discrepancies, reflected in uncertainty quoted in table; e.g.:
- discrepancies exceed individual experimental errors;
- average of a large number of experiments including some discrepant values;
- some discrepant values, but majority within uncertainty
notes: explanations to values shown

f) The percent contribution to the total FP afterheat as a function of cooling time is indicated as follows:

——— 1-2%
▬▬ 2-5%
▬▬ 5-10%
▬▬ ≥10%
maximum of 5 cases calculated by C. Devillers (appendix to RP 4)
------ precursor only

g) 3 isomeric states have been observed with half-lives 48 ± 3 s, 83 ± 3 s and 100 ± 3 s [3]. According to Devillers' calculation the 83 s isomer is indicated as ground state, whereas in [1] the 48 s isomer is given as ground state.

h) Half-life of ≲ 0.3 s reported in [14]. Therefore experimental uncertainty questionable.

i) No uncertainty given for this value; uncertainty deduced from other measurement quoted in [14].

k) [1] gives 2.5 m for ^{100}Nb. In [15] 1.5 ± 0.3 s ($\pm 20\%$) are reported for ^{100}Nb, following decay of ^{100}Zr. For half-life assignment see discussion in [15].

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