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## U.K. NUCLEAR DATA PROGRESS REPORT for the period, January to December 1977

Editor: D.B. Syme Nuclear Physics Division AERE Harwell, Oxfordshire June 1978

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#### PREFACE

This report is prepared at the request of the United Kingdom Nuclear Data Committee and covers the period from January to December 1977. Contributions on nuclear data topics are included from Harwell, Winfrith, Dounreay, MOD Aldermaston, Imperial College, the National Physical Laboratory, the National Radiological Protection Board and the Universities of Birmingham, Edinburgh, Glasgow and Manchester.

As before, contributions from "chemical nuclear data" are grouped under that heading but other reports are presented under a laboratory heading. Where the work is clearly relevant to requests in WRENDA 76/77 (INDC(SEC) 55/URSF) the request numbers are given after the title of the contribution. A CINDA type index is included.

The text of a lecture given to this year's Nuclear Data Forum and entitled "The Future of Fission Reactors" is reproduced at the front of the document.

#### COMMITTEE ACTIVITIES

#### United Kingdom Nuclear Data Committee (Chairman: Dr. J.E. Lynn)

The UKNDC met twice during the period of this report, and continued its role of co-ordinating work on nuclear data in the U.K. and providing the formal link with other national and international nuclear data committees and organisations. Detailed nuclear data needs were dealt with by the five sub-committees, summary reports from which are given below.

An International Conference on Neutron Physics and Nuclear Data for Reactors and other Applied Purposes is to be held at Harwell in September 1978. This will be the first in a series of triennial European conferences which will be held alternately with similar conferences in the USA and the USSR to provide annual meetings.

The eleventh Nuclear Data Forum was held in December at Imperial College of Science and Technology. The theme for the morning session was data for reactor fuel cycles and the main talks, on the future of fission reactors and the physics of alternative fuel cycles, were received with great interest. We have been fortunate to secure the text of one of these and it is reproduced elsewhere in this report. The later sessions were devoted to the experimental results and calculational methods of nuclear data. The meeting was attended by about 80 scientists from the UKAEA, from government and the nationalised industries and from the universities.

#### Neutron Sub-committee (Chairman: Dr. J.E. Lynn)

The committee did not meet during the period covered by this report. Nuclear Incineration Sub-committee (Chairman: Dr. M.G. Sowerby)

There was no committee meeting during the period covered by this report.

#### Fusion Sub-committee (Chairman: Dr. C.A. Uttley)

The committee did not meet during the period covered by this report. Chemical Sub-committee (Chairman: Mr. J.G. Cuninghame)

Chemical nuclear data experiments and evaluations are coordinated by the Chemical Nuclear Data Committee. This committee, which is also the Chemical Sub-Committee of the UKNDC, is made up of measurers, evaluators and users of chemical nuclear data and is able to advise on measurements and to consider in detail reports of work on the compilation and evaluation of data in this field. The preparation of the CNDC nuclear data file is under the control of the Data File Sub-Committee (DFSC) whose new chairman is Mr. B.S.J. Davies (CEGB, BNL). The main committee held two meetings

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and the sub-committee three meetings in 1977. The request list was revised early in the year (CNDC(77) Pl) and is due to be published in conjunction with the 'Physics' list. Work is in progress on very nearly all items on the list but often on a long time scale and with little prospect of early completion. Data File work, however, has now reached the stage where data is available to the user.

The efforts of the committee to reduce the duplication of work have been successful and the objective of effective international cooperation in the data field is being pursued.

#### Bio-Medical Sub-Committee (Chairman: Mr. J.A. Dennis)

The Bio-Medical Sub-Committee held its fourth meeting on the 3rd October 1977. The Sub-Committee noted that a recommendation had been made to disband the INDC Sub-Committee on Non Energy Applications although it was thought that the INDC might recommend holding Specialists' Meetings on Nuclear Data for Biomedical Needs. A view was expressed that such meetings should be related to the CENDOS project (Collection and Evaluation of basic data relevant to Neutron Dosimetry) sponsored by Euratom.

There was some disappointment that there had been no official response to the request list for nuclear and atomic data measurements for use in the biomedical field, prepared by the Sub-Committee, and a decision was made to circulate the list internally within the U.K.

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The draft of an article on available sources of nuclear and atomic data for biomedical applications was discussed and amendments suggested.

#### THE FUTURE OF FISSION REACTORS

#### R H Flowers

Chemical Technology Division, AERE, Harwell

Lecture given at the Nuclear Data Forum, Imperial College, 12th December 1977 Introduction

I must explain first of all that the title 'The Future of Fission Reactors,' is possibly misleading. Interesting though the subject is, I do not intend to embark upon a discussion of the 'energy gap' and the suitability of fission reactors to fill it; that discussion has been well aired on other occasions. I would like instead to look briefly at the state of nuclear data in the context of current civil reactor strategy and to suggest some directions in which nuclear data requirements may be moving in order to satisfy the next generation of fission reactors. My view of the nuclear data scene is, I should say, that of a chemist with a long association with nuclear reactor design and operation - in other words a user rather than a measurer.

The present civil nuclear power strategy in the world is, roughly speaking, one of operating thermal reactors to fission about 0.5% of the uranium atoms in natural ore. The atoms fissioned are mainly  $^{235}$ U, but not entirely, since depending upon reactor type around 10% of the fissions may be of  $^{238}$ U, via the intermediate species  $^{239}$ U,  $^{239}$ Np and  $^{239}$ Pu. The strategy is then to reprocess spent fuel in order:-

(a) to recover  $^{239}$ Pu to fuel further thermal or fast reactors

(b) to recover  $\begin{array}{c} 235 \\ U \end{array}$  and  $\begin{array}{c} 238 \\ U \end{array}$  to fuel further thermalor fast reactors,

usually after enrichment in a diffusion plant

(c) to put fission products in a disposable form.

Nuclear data have of course been fundamental in reaching the position which we hold today in both thermal and fast reactor construction and operation capability. The principle applications are:-

(1) Core physics

(2) Fuel chemistry

- (3) Reactor thermal performance
- (4) Radiological protection and environmental impact
- (5) Reprocessing
- (6) Safeguards

Conferences held by the IAEA at Bologna in 1973<sup>(1)</sup> and Petten in 1977<sup>(2)</sup> made a careful evaluation of a large section of this field, namely the requirements for Fission Product Nuclear Data and the status of measurements of those data. The whole area of nuclear data for these applications seems to me to be very well organised on an international basis, at least in the context of the currently accepted reactor strategy.

#### Data situation for the present strategy

Taking the six headings in turn:-

#### (1) Core physics

The key data in core design are fission cross-sections of the fissile actinides, delayed neutron precursor yields, and neutron capture cross-sections in actinides, fission products, moderators and core constructional materials. Fission product yields and decay schemes are also important in calculating inventories.

For thermal reactors, Rowlands concluded in his paper to the Petten conference that fission product and delayed neutron precursor yield data are now reasonably satisfactory for core physics purposes ( $\pm 10\%$ ) - including the necessary temperature coefficients of capture cross-sections. I believe the actinide capture and fission crosssections, at least for the important <sup>235</sup>U and <sup>239</sup>Pu isotopes are also sufficiently well known.

For the fast reactor, with burn-up up to 10%, Rowlands concluded that the fission product data situation is now satisfactory ( $\pm$  25% on  $\alpha$  and inventory), apart possibly from the energy dependence data on some fission products, for the calculation of sodium void and Doppler effects, and some refinement of delayed neutron data. Actinide capture and fission cross-sections must by now I think be adequately known for the LMFBR core physics purposes.

#### (2) Fuel chemistry

The reactor chemist requires to know the chemical composition of the fuel as burn-up proceeds because this determines corrosion of the cladding, tendencies to phase changes or perhaps swelling of the fuel and the gas pressure built up to Kr, Xe and perhaps CO or  $H_2$ .

This is a question of knowing the fission product yields, decay schemes and neutron capture cross-sections well enough to allow calculation of an inventory to  $\pm$  5%-10% as a function of rating and burn-up and reactor type. There are few deficiencies here, provided we are interested in thermal or LMFBR type neutron spectra and in burn-ups

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#### of up to 3% and 10% respectively.

Table 1 shows an example of the importance of fission product inventory, in this case in the calculation of gas pressure inside a carbon-coated particle.

Table 1

Fission Product Inventory and Oxygen Balance in a Coated UO<sub>2</sub> Particle

Initial UO<sub>2</sub> mass = 0.0025g Enrichment = 8%

a - · ·

Burn-up 10% FIMA Dwell = 1000 days

. .

	Element	Mass x 10 <sup>3</sup> (g)	Oxidised to	$\begin{array}{c} \text{Mass } 0_2 \times 10^3 \\ (g) \end{array}$
	Xenon	0.0334	-	-
' i	Zirconium	0.0219	Zr0 <sub>2</sub>	0.0076
	Neodymium	0.0224	NdO <sub>2</sub>	0,0050
	Molybdenum	0.0210	-	-
	Cerium	0.0171	CeO <sub>2</sub>	0,0039
	Ruthenium	0.0148	-	· · · -
	Caesium	0.0174	<del></del>	·
	Barium	0.0087	BaO	0.0010
	Palladium	0.0082	-	-
	Lanthanum	0.0077	La0 <sub>2</sub>	0.0018
	Technetium	0.0051		2 - 2 - <sup>1</sup> - <del>1</del>
	Strontium	0.0054	Sr0	0,0010
	Tellurium	0.0036	· -	· · ·
	Rubidium	0.0020		-
	Iodine	0.0018	. – .	19. – S
	Rhodium	0.0028	. –	-
	Promethium	0.0012	PmO <sub>2</sub>	0.0003
	Praesodymium	0.0073	Pr02	0.0016
	Silver	0.0006	-	-
	Krypton	0.0022	-	-
	Samarium	0.0040	SmO <sub>2</sub>	0.0008
	TOTAL	0.21		0.023
•	Carbon	· · · · · · · · · · · · · · · · · · ·	CO	0.007
:	Uranium	1.99	U0 <sub>2</sub>	0.27
	GRAND TOTAL	2.20	_	0.30

#### (3) <u>Reactor thermal performance</u>

Apart from the key figure for MeV per fission of the uranium and plutonium isotopes, which is well enough known for the purposes of fuel management, the most important

data in this area are those which determine the thermal power of a core as a function of time after shutdown. M. Devillers (CEA Saclay) reviewed this field at the Petten meeting this year.

Calculation of fission product decay power should be accurate to 5-10% over a period 0-8 hours for thermal reactor operation, and 0-24 hours for fast reactors. The integral decay energy is sufficient for very short times (less than 1 minute) and so nuclear data of extremely short half-life nuclides are not required.

Inventories and decay energies are the key data - with inventories as usual requiring a knowledge of fission yields, decay schemes and neutron capture cross-sections. Devillers concluded:-

(a) that yields and decay schemes are not responsible for significant errors

(b) that decay energies are limiting the accurancy obtainable,

but nevertheless he found that the 5-10% required accuracy in total thermal power was achievable for the present reactor types.

#### (4) Radiological Protection

In operating a reactor, attention has to be given to gaseous and liquid discharges and to gamma-dose to the station staff. The analysis of these problems involves a study of fission product release from fuel elements, leakage of coolants and of the behaviour of radionuclides in animals and in the environment - features which do not concern us here. However, the inventory of actinides and fission products again forms the starting point of all such assessments. The usual procedure is to evaluate the behaviour of a few key nuclides, chosen on the basis of their toxicity, mobility and concentration. For example  $^{137}$ Cs is a good control isotope on which to assess the problems of gamma-dose from leaking fission products in a reactor circuit. Fig 1 shows the logic which is applied, in this instance to relate HTR coated particle fuel quality to the y dose from boiler maintenance.

The end point in these calculations is a gamma-dose rate to staff or an inhalation or ingestion rate by persons at the boundary fence. Since the dose limits are couched in much less precise terms than the accuracy of most nuclear data (factors of 2 are not usually important) the necessary inventory calculations can be made without difficulty, except for a few special cases. <sup>3</sup>H yield in fast reactors still seems to be in need of refinement, and the products of neutron capture on some fission products and actinides may also be uncertain to a significant extent.

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## <sup>137</sup>Cs as a Maintenance Hazard





(5) Reprocessing

Present reprocessing plants are sensitive to inventory of actinides and fission ...
products from the points of view of:-

- (a) composition of insoluble residues of Mo, Tc, Ru, Pd, Pu
- (b) heat generation in fuel during handling
- (c) gamma-shielding required on the plant
- (d) Neutron emitters in spent fast reactor fuel
- (e) chemical composition of stored wastes

McKay reported at Petten that, for the present reactor strategy, fission product nuclear data are sufficiently accurately known for these purposes; the position with

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includes is similar with the possible exception of significant uncertainties in the inventory of the neutron emitters  $^{242}$ Cm and  $^{244}$ Cm in high burn-up fast reactor fuel.

(6) <u>Safeguards</u>

The problems of analysing for fissile nuclides by non-destructive methods, even in irradiated fuel or in process liquors of a reprocessing plant, were reviewed by N.J. Maeck at the Petten 1977 meeting. He concluded that although most of the neutron interrogation methods relied upon direct comparison with standards, there is room for improvement in a few selected fission yields, capture cross-sections and gamma-intensities. Also in this paper Maeck points out that refinement of data on the neutron cpature of particular fission products, especially at non-thermal neutron energies, would allow better accuracy of fast-reactor burn-up measurements.

The overall conclusion is that, whilst data measurement should continue in support of our present reactor strategy, progress in civil nuclear power introduction is not rate-limited by the shortcomings.

#### 7. Future Trends

During the past five years the rate-limiting step in civil power introduction has shifted from reactor design to other factors in the out-of-reactor part of the fuel cycle. This does not mean that the present thermaland fast reactor designs are perfected, but it does show that public apprehension has shifted from the safety of reactors to the safety of fuel materials outside the reactor.

I would say that there are four major obstacles to rapid nuclear power growth, two of them real and two of them partly the result of a false public perception of the technical facts. They are:-

(a) the fear of encouraging nuclear weapons proliferation

- (b) the complexity and therefore high capital cost of stations which discourages
  - load following applications
- (c) the limited availability of uranium ore in many parts of the world

(d) the fear of waste products requiring supervision for many thousands of years. The first of these relates to plutonium, which is formed in our thermal reactors and is the basis of our fast reactor programme. The second results from the high standards of safety and reliability set by the nuclear power industry. The third is a fact; it is worth remembering that the probable resources of  $^{235}$ U are equivalent only to the oil reserves and that, unless  $^{238}$ U or  $^{232}$ Th is fissioned effectively, many countries would very soon lose interest in nuclear power. Table 2 shows this point in

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terms of tonnes of coal equivalent. The fourth relates to the occurrence of artificial nuclides <sup>239</sup>Pu, <sup>243</sup>Cm, <sup>243</sup>Am, <sup>239</sup>Np in the spent fuel and consequently in waste streams from the industry. Their disposal is not actually such a difficult and novel task as is popularly believed but that does not immediately remove the apprehension.

#### Table 2

Resources	of	Energy	'Captial'
	_		

		Quantity		
Resource	Wo	rld	UK	Comment
	Tonnes	TCE	TCE	
Coal	10 <sup>13</sup>	10 <sup>13</sup>	10 <sup>11</sup>	1.5x10 <sup>8/</sup> y in UK by 1985
011	3x10 <sup>11</sup>	5x10 <sup>11</sup>	~ 10 <sup>10</sup>	1.8x10 <sup>8</sup> /y " " " "
Gas	5x10 <sup>15</sup> (cu ft)	2x10 <sup>11</sup>	. 5x10 <sup>9</sup> ,	8x10 <sup>7</sup> /y " " " "
Uranium	7 x 10 <sup>4</sup>	$1.4 \times 10^{11}$	-	500ppm ores
Uranium-238	10 <sup>7</sup> ,	2x10 <sup>13</sup>	4x10 <sup>10</sup>	500ppm ores
Thorium-232	10 <sup>7</sup>	2x10 <sup>13</sup>	-	not well explored
Lithium	10 <sup>7</sup>	2x10 <sup>13</sup>	_	to produce <sup>3</sup> H for D+T
Geothermal	-	10 <sup>20</sup>	-	3x10 <sup>13</sup> W in world
Lunar	-	5.10 <sup>18</sup>	_ •	3x10 <sup>12</sup> W " "
Uranium in sea	4x10 <sup>9</sup>	8x10 <sup>15</sup>	-	unlikely to be in time
Deuterium in sea	2x10 <sup>14</sup>	-10 <sup>20</sup>		feasibility unexplored

In my view, the current civil nuclear power strategy of thermal reactors providing fissile plutonium to launch LMFBRs for the beginning of the next century <u>is</u> a viable means of utilising our  $^{238}$ U resources, and President Carter's April 1977 call for a world reappraisal of the problems which it raises will show that those problems can be handled without a revision of the whole strategy and the decades of delay which that would incur.

For example:-

(a) in the reprocessing of either thermal or fast reactor fuel, we can change the conventional process in which plutonium is separated in a pure state, to one in which only a U/Pu mixture suitable for reactor fuel is separated.

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(b) before committing reprocessing wastes to underground disposal we could separate chemically the actinide elements and incorporate them in reactor fuel; this would remove the preceived problem of burial of very long-lived nuclides.

What then of the prospects for new fuel cycles in the long term - fuel cycles to take over from the present ones during the next century because they are:-

- (a) more difficult to bend towards weapons production
- (b) better suited to use our  $^{238}$ U and  $^{232}$ Th resources
- (c) lest we forget cheaper.

There is no time for new kinds of thermal reactor having conversion ratios around 0.5 (like existing types) because such reactors can at best utilise an amount of  $^{238}$ U or  $^{232}$ Th equivalent to the  $^{235}$ U which we feed to them. The future must lie with reactors of conversion ratio around unity or higher; whether they operate in a thermal or epithermal neutron spectrum is less important. The new reactors must be simpler to build (so that their capital cost is reduced), they should avoid the handling of high concentration fissile actinides (to remove them even further from the weapons arena), and if they can be made to burn their own long-lived radioactive products so much the better.

#### 8. Nuclear Data for the Future

High conversion ratio thermal reactors, taking advantage of the high neutron yield from <sup>233</sup>U in a thermal spectrum, might have a part to play in bringing <u>some</u> of the world's <sup>232</sup>Th resources into use, probably in countries where CANDU reactors are established. There is scope for improving knowledge of some of the many reaction rates leading to the formation of <sup>232</sup>U from <sup>232</sup>Th and <sup>230</sup>Th - this <sup>232</sup>U isotope decays through to the 2.8 MeV  $\gamma$ -emitter <sup>208</sup>Tl. It has been suggested that use of <sup>238</sup>U as a diluent of the fissile feed to a Th converter reactor would avoid the handling of highly pure <sup>235</sup>U or <sup>233</sup>U - the physics of such complex cores needs evaluation to see whether there is indeed any advantage, bearing in mind that the <sup>238</sup>U will breed Pu. I must leave the physicists to say whether present actinide data are sufficient for that task.

Finally I believe that we should be now looking more seriously for a second generation <u>fast</u> reactor which will show that there is scope in nuclear fission for a gradual evolution of plants which meet and overcome the four obstacles to a greater degree than can be achieved through refinements to the LMFBR. It is a responsiblity of

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nuclear data experts to foresee such developments as the GCFR, the thorium burning fast reactor and very high burn-up fuels, so that they can prepare the way for very advanced reactors making good use of our  $^{238}$ U and  $^{232}$ Th in the long term.

Ultimately, I suggest, we should aim at a gas-cooled fast reactor with a neutron economy good enough to allow it to burn replacement fuel of natural uranium or thorium, once given its initial fissile inventory. Evolution in that direction would suggest that fission yields, and neutron capture and fission cross-sections, will be required for harder neutron spectra than found in the LMFBR and that multiple neutron capture on individual fission products will become more significant at the much higher neutron doses involved.

There are, of course, materials problems in such a reactor but that would be the subject for a different meeting.

- 1. IAEA-169 Fission Product Nuclear Data (FPND) Vol. 1. Proceedings of a Panel on Fission Product Nuclear Data organised by the International Atomic Energy Agency and held in Bologna, 26-30 November 1973.
- Second IAEA Advisory Group Meeting on Fission Product Nuclear Data (FPND) 5-9 September 1977 in Petten. Proceedings to be published.

#### CINDA LISTINGS

ELEMENT			TUDE	ENE	RGY	D	OCUME	NTATIO	N	TAP	COMMENTS	
S A	А	QUANTIT		MIN.	MAX.	REF	VOL	PAGE	DATE	LAD	CONTIENTS	
н	1	TOTAL XSECT	THEO	-	5.0 +7	UKNDC	P88	112	6/78	UK	DIMBYLOW Optical model fits to total xsec	
Li	7	N,N ALPHA	EXPT- PROG	2.7 +6	1.4 +7	UKNDC	P88	60	6/78	HAR	UTTLEY+	
С	12	TOTAL XSECT	THEO	-	5.0 +7	UKNDC	P88	112	6/78	UK	DIMBYLOW Optical model fits to total xsec	
N	14	TOTAL XSECT	THEO	-	5.0 +7	UKNDC	P88	112	6/78	UK	DIMBYLOW Optical model fits to total xsec	
0	16	TOTAL XSECT	THEO	-	5.0 +7	UKNDC	P88	112	6/78	UK	DIMBYLOW Opticalmodel fits to total xsec	
F	19	ACTIV- ATION	EXPT- PROG	FISS	-	UKNDC	P88	71	6/78	HAR	HOOTON+	
AL	27	ACTIV- ATION	EXPT- PROG	FISS	_	UKNDC	P88	71	6/78	HAR	HOOTON+	
Si	NAT	ACTIV- ATION	EXPT- PROG	FISS	-	UKNDC.	P88	71	6/78	HAR	HOOTON+	
CR	NAT	N,ALPHA	EXPT- PROG	-	1.4 +7	UKNDC	P88	60	6/78	HAR	COOKSON Work planned	
FE	NAT	N,GAMMA	EXPT- PROG	1.0 +2	1.0 +6	UKNDC	P88	38	6/78	HAR	GAYTHER+ L.L.S. relative to gold 589mb at 30 keV	
FE	NAT	RESON PARAMS	EXPT- PROG	0.0 +0	2.3 +5	UKNDC	P88	47	6/78	HAR	MOXON+ Shape fitting to transmission data	
FE	NAT	DIFF ELASTIC	EXPT- PROG	-	1.6 +7	UKNDC	P88	121	6/78	EDG	GALLOWAY+ Angular dist- tribution of scattered polarised neutrons	
FE	NAT	N,ALPHA	EXPT- PROG	-	1.4 +7	UKNDC	P88	60	6/78	HAR	COOKSON <sup>+</sup> Work planned	
FE	54	N, 2N	EXPT- PROG	1.4 +7	1.9 +7	UKNDC	P88	107	6/78	NPL	RYVES <sup>+</sup> data to be present in Metrologia July 78.	
FE	56	N,proton	EXPT- PROG	1.4 +7	1.9 +7	UKNDC	P88	107	6/78	NPL	RYVES+ data to be present in Metrologia July 78.	
Ni	NAT	N,GAMMA	EXPT- PROG	1.0 +2	1.0 +6	UKNDC	P88	38	6/78	HAR	GAYTHER+ L.L.S. relative to gold 589mb at 30 keV	
Ni	NAT	RESON PARAMS	EXPT- PROG	0.0 +0	3.0 +5	UKNDC	P88	52	6/78	HAR	SYME+ Shape and area fits to transmission data	

#### CINDA LISTINGS

ELEMENT				ENE	RGY	D	OCUME	NTATIO	N		COMPATE	
S	А	QUANTITY	TYPE	MIN.	MAX.	REF	VOL	PAGE	DATE	LAB	COMMENIS	
Ni	NAT	N,ALPHA	EXPT- PROG	_	1.4 +7	UKNDC	P88	60	6/78	HAR	COOKSON work planned	
Ni	58	RESON PARAMS	EXPT- PROG	0.0 +0	5.0 +5	UKNDC	P88	52	6/78	HAR	SYME+ Shape and area fits to transmission data	
Cu	NAT	DIFF ELASTIC	EXPT- PROG	-	1.6 +7	UKNDC	P88	121	6/78	EDG	GALLOWAY+ angular dist- tribution of scattered polarised neutrons	
Cu	63	N, 2N	EXPT- PROG	1.61+7	1.9 +7	UKNDC	P88	107	6/78	NPL	RYVES+ data to be pres- ent at Metrologia July 78	
Cu	65	N,P	EXPT- PROG	1.61+7	1.9 +7	UKNDC	P88	107	6/78	NPL	RYVES+ data to be present in Metrologia July 78	
Cu	65	N,N ALPHA	EXPT- PROG	1.61+7	1.9 +7	UKNDC	P88 <sup>-,</sup>	107	6/78	NPL	RYVES+ data to be present at Metrologia July 78	
Cu	65	N, 2N	EXPT- PROG	1.61+7	1.9 +7	UKNDC	P88	107	6/78	NPL	RYVES+ data to be present in Metrologia July 78	
Y	89	IN- ELASTIC Y	EXPT- PROG	-	FAST	UKNDC	P88	127	6/78	GLS	CRAWFORD+ Ge-Li detector used for in- elastic gamma spectra	
I	127	DIFF. ELASTIC	EXPT- PROG	-	1.6 +7	UKNDC	P88	121	6/78	EDG	GALLOWAY+ angular dist- ribution of scattered polarised neutrons	
W	NAT	DIFF. ELASTIC	EXPT- PROG	-	1.6 +7	UKNDC	P88	121	6/78	EDG	GALLOWAY+ angular dist- ribution of scattered polarised neutrons	
W	NAT	DIFF. ELASTIC	EXPT- PROG	-	2.9 +6	UKNDC	P88	123	6/78	EDG	BEGUM+ Sig polarisation and angular distribution	
нв	NAT	DIFF. ELASTIC	EXPT- PROG	-	1.6 +7	UKNDC	P88	121	6/78	EDG	GALLOWAY+ angular distribution of scatt- ered polarised neutrons	
T1	NAT	DIFF. ELASTIC	EXPT- PROG	-	2.9 +6	UKNDC	P88	123	6/78	EDG	BEGUM+ Sig polarisation and angular distribution	
Рb	NAT	DIFF. ELASTIC	EXPT- PROG	· _	1.6 +7	UKNDC	P88	121	6/78	EDG	GALLOWAY+ angular distribution of scatt- ered polarised neutrons	
РЪ	NAT	DIFF. ELASTIC	EXPT- PROG	_	2.9 +6	UKNDC	P88	123	6/78	EDG	BEGUM+ Sig polarisation and angular distribution	

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#### CINDA LISTINGS

ELEMENT		OULINETEN	TYDE	ENE	RGY	DOCUMENTATION					CONCENTS	
S	А	QUANILIY	TYPE	MIN.	MAX.	REF	VOL	PAGE	DATE		COMMENTS	
Bi	209	DIFF. ELASTIC	EXPT- PROG	-	2.9 +6	UKNDC	P88	123	6/78	EDG	BEGUM+ Sig Polarisation and angular distribution	
U	NAT	DIFF. ELASTIC	EXPT- PROG	-	2.9 +6	UKNDC	P88	123	6/78	EDG	BEGUM+ Sig Polarisation and angular distribution	
U.	NAT	TOTAL XSEC	EXPT- PROG	4.0 +0	2.0 +5	UKNDC	P88	28	6/78	HAR	HASTE+ UO2 total x- section at 3 temps.	
U	233	N, FISSON	EXPT- PROG	1.0 +5	2.0 +7	UKNDC	P88	33	6/78	HAR	JAMES+ Ratio to U-235	
U	235	TRITION YIELD	EXPT- PROG	THER	-	UKNDC	P88	90	6/78	HAR	McKEAN+ Trition yields per fission	
U	235	SPECT FISS N	EXPT- PROG	1.0 +5	_	UKNDĊ	P88	59	6/78	HAR	ADAMS+ Analysis in progress	
U	238	RESON PARAMS	EXPT- PROG	6.7 +0	1.9 +2	UKNDC	P88	• 24	6/78	HAR	HASTE+ Multi-level shape fits to trans- mission data	
Pu	239	N, FISSION	EXPT- PROG	1.0 +5	2.0 +7	UKNDC	P88	33	6/78	HAR	JAMES+ Ratio to U-235	
Pu	241	HALF LIFE	EXPT- PROG	-	-	UKNDC	<b>Р́8</b> 8	97	6/78	HAR	CROUCH Half Life	
Am	241	N,GAMMA	EXPT- PROG	1.0 +2	1.0 +6	UKNDC	P88	35	6/78	HAR	GAYTHER+ Capture cross- section energy depen- dence.	
Am	241	N, FISSION	EXPT- PROG	5.0 +2	1.0 +4	UKNDC	P88	35	6/78	HAR	GAYTHER+ relative to U-235 fission	
Am	241	RES INT	EXPT- PROG	FAST	-	UKNDC	P88	96	6/78	HAR	GLOVER+ CM 242 yield xsec = 1.275+0.013b	
Am	243	RES INT	EXPT- PROG	FAST	-	UKNDC	P88	96	6/78	HAR	GLOVER+ CM 244 yield xsec = 1.51 b	

#### NUCLEAR PHYSICS DIVISION, A.E.R.E., HARWELL

(Division Head; Dr. B. Rose.)

#### EDITORIAL NOTE

Since the results obtained from the various machines are not easily classified according to the energy of the charged beams, individual research items are labelled with a single letter indicating on which machine the experiments were performed. These labels are as follows:

Cockcroft-Walton Generator (G. Dearnaley)	A
3 MV pulsed Van de Graaff Generator IBIS (A. T. G. Ferguson)	В
6 MV Van de Graaff Generator (A. T. G. Ferguson)	С
14 MeV Tandem Generator (J. M. Freeman)	D
45 MeV Electron Linac (J. E. Lynn)	Ε
Variable Energy Cyclotron (Chemistry Division)	G
Synchrocyclotron (C. Whitehead)	Ħ

The running analysis for the various machines operated by the division are presented as far as possible in a uniform format, but some differences exist in the way in which the scheduling is arranged, and machines such as the Electron Linac can accommodate several experiments simultaneously.

Where the work is clearly relevant to requests in WRENDA 76/77 (INDC(SEC)-53/URSF) request numbers are given after the title of the contribution.

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#### NUCLEAR DATA AND TECHNOLOGY FOR NUCLEAR POWER

#### E. The new machine for the electron linac laboratory (J. E. Lynn, M. S. Coates, P. P. Thomas and B. P. Clear)

The building programme on the project has essentially kept to the schedule envisaged originally in PR/NP 22, p 9. All construction work (see PR/NP 22, p 10) is expected to be finished early in 1978 including a new experimental hall which is attached to the east side of the condensed-matter cell. This hall, which was not included in the original project and which has been separately funded, will contain some of the experimental equipment to be used on the condensed-matter cell neutron beam lines. All of this work has been carried out by Chivers Ltd. to the design and specification of the Building Design and Construction Group of Engineering Division.

Production of the new linac by Radiation Dynamics Ltd. of Swindon has suffered a delay of some months and the present R.D.L. estimate of the delivery date of the machine to Harwell is late summer of 1978. Works tests have been successfully made on the first of the four modulator units and a beam has been accelerated down the first of the eight accelerating sections. Beam tests using a maximum of only two sections in series can be made in the R.D.L. works and the final eight section assembly will be tried after installation at Harwell, although all sections will have been run at some time in the R.D.L. test bed.

All of the four klystron valves needed for the full accelerator have been produced by Thomson-CSF (UK) Ltd. and are ready when required.

The production of the electron beam handling magnet and quadrupole elements is well advanced and a significant amount of installation has been done already. It is anticipated that all of this equipment will be ready for use when needed.

Neutron source targets for the fast-neutron and condensed-matter cells have been designed. Basically each of the first choice targets consists of a series of water cooled clad natural uranium plates contained in a pressure vessel. The condensed matter target (50 kW(e) power dissipation) has Zircaloy 2 as the cladding material with an Inconel container. The fast neutron target (30 kW(e) power dissipation) has tantalum cladding and a beryllium container. Production and other technical difficulties however make it doubtful whether clad uranium plates can be made to meet the required specifications. For this reason two additional targets each having unclad tantalum plates have been designed and these will be available by mid-summer 1978. Such a tantalum target has a

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lower neutron yield by a factor of ~2 than one made from natural uranium.

At this stage it is hoped that the linac will be ready for experimental use at Harwell by the end of 1978.

A description of the linac project, including details of the expected linac performance, was given at the Fourth National Soviet Conference on Neutron Physics<sup>(1)</sup>.

(1) M. S. Coates, P. P. Thomas, B. P. Clear, R. N. Sinclair and J. E. Lynn, A new electron linear accelerator at A.E.R.E., Harwell, paper presented at the IV National Soviet Conference on Neutron Physics, Kiev, USSR, April 1977

#### E. Resonance parameters of <sup>238</sup>U below a neutron energy of 200 eV (T. J. Haste, <u>M. C. Moxon and J. E. Jolly)</u> [Relevant to request numbers: 852, 854-8]

The determination of the effective temperature from the neutron energy dependence of the transmission of heated samples of  $UO_2$  requires accurate values of the resonance parameters.

A consideration of all the available measurements (1-16) of the parameters of the s-wave resonances for  $^{238}$ U up to a neutron energy of 189 eV shows that there is reasonable agreement on the different determinations of the resonance energies. Measurements for given resonances of both the neutron and radiation widths show fluctuations well outside the limits of the uncertainty. Two contributing factors towards these discrepancies could be (a) non-uniformity in the thickness of the thin samples and (b) invalid assumptions made in the analysis of the data.

To overcome the problems of the thin sample uniformity, neutron transmission measurements were carried out on three conentrations of uranyl nitrate solution in heavy water prepared by D. H. McGahan of the Chemistry Division at Harwell. The solutions were contained in parallel-sided silica vessels of the type used in calorimetric measurements.

The neutron transmission measurements were carried out on the 14 m small sample flight path of the booster target of the Harwell 45 MeV linac before the machine closed down at the end of 1976.

The resonance parameter analysis of these data, together with the transmission data obtained at different temperatures with the  $UO_2$  samples was carried out using the program REFIT <sup>(17)</sup>. This program uses the R-matrix multi-level formalism to describe the nuclear cross-section. Doppler broadening of the cross-sections is carried out using the ideal gas model with an effective temperature which can be adjusted. The calculated

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transmission is convoluted with the resolution function for comparison with the experimental data.

Figure 1 shows a least squares fit to the transmission data of the three uranyl nitrate samples for the 20.9 eV resonance. Here the resonance energy, the neutron width and a renormalisation factor for each sample were taken as adjustable parameters. Figure 2 shows the fit to the UO<sub>2</sub> samples for the 20.9 eV resonance obtained by adjusting the resonance energy, the radiation width and renormalisation factor, assuming the value of the neutron width of 10.17 meV obtained from the solution data. The neutron widths obtained from fits to the uranyl nitrate data for all the s-wave resonances below 189 eV







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were found to be almost independent of the radiation width when it was varied between 20 and 30 meV. The radiation widths for the resonances at 6.6, 20.9, 36.9 and 66.0 eV were obtained from fits to the transmission data on the thick UO<sub>2</sub> samples, assuming the values of the neutron widths from the fits to the solution data.

Table 1 lists the values of the s-wave resonance parameters from the least squares fits to the present data up to an energy of 189 eV. The quoted uncertainties are one standard deviation and are the statistical uncertainties obtained from the fits to the data. A quadratic sum of the co-variance terms in the uncertainty matrix between the parameters being fitted appears at present to be between 1 and 2 times the statistical uncertainties. Further investigation will be made here.

#### Table 1

<sup>&</sup>lt;sup>238</sup>U s-wave resonance parameters obtained from least squares fits to transmission measurements carried out on the Harwell 45 MeV linac

Resonance	Statistical	Neutron	Statistical	Radiation	Statistical
energy	uncertainty	width	uncertainty	width	uncertainty
(eV)	(eV)	(eV)	(eV)	(eV)	(eV)
6.6680 20.8773 36.6916 66.0556 80.7616 102.5940 116.929 145.563 165.187 189.789	0.00030 0.00080 0.0013 0.0177 0.0048 0.0067 0.054 0.019 0.013 0.020	1.5070 10.17 34.41 25.64 + 1.794 *69.73 +25.18 + 0.814 + 2.931 *183.63	0.0080 0.10 0.41 0.66 0.058 0.52 0.18 0.052 0.157 1.49	23.54 22.44 24.03 23.46 (24.0) (24.7) (23.0) (23.5) (23.5) (23.5) (23.4)	0.53 0.43 0.87 0.50

\* weighted mean values from the UO<sub>2</sub> data and solution data

 $^{\rm +}$  the neutron width obtained from fits to the UO  $_2$  data assuming the radiation width given in brackets

In general the fitted values of the resonance energies are in good agreement with the weighted mean values of all the previous measurements. However our values of the neutron and radiation widths are in better agreement with the latest values from Geel<sup>(16)</sup>, than with the evaluated values of all previous measurements. In the case of the first two resonances, both Geel<sup>(16)</sup> and our values differ from the previously accepted values. In the case of the 6.668 eV resonance our value of the radiation width is smaller than the weighted mean including the Geel results. The low weighted mean value of 24.59  $\pm$  0.50 for the radiation width of the 6.668 eV resonance was only obtained by increasing the

error from  $\pm$  0.4 to  $\pm$  10 meV on the value of 27.2 obtained by Jackson and Lynn<sup>(8)</sup> to take into account possible effects of non-uniformity in thickness of the thin samples used in these measurements. The neutron width of the 20.9 eV resonance of 10.17  $\pm$  0.10 meV is in good agreement with the Geel result of 10.2  $\pm$  0.1 meV but in disagreement with previous values which tend to be much lower (a value of 8.5 meV being quoted in BNL 325<sup>(18)</sup>). The only other resonance showing a large discrepancy with previous measurements is the one at 189.8 eV where we obtained a value of 183.6  $\pm$  1.5 meV, larger than the Geel value of 164.0  $\pm$  3.0 meV and much larger than the weighted mean of 156.3  $\pm$  3.2 meV.

We have almost completed our analysis of these data and at present we are investigating the uncertainties in the parameters. A detailed report of this work will appear shortly.

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(18)	S. F. Mughabghab and D. I. Garber, BNL-325, 3rd Edition, Vol. 1 (June 1973)

÷27-

E. Doppler broadening studies on uranium dioxide (T. J. Haste and M. G. Sowerby) [Relevant to request numbers: 852, 854, 856]

Neutron transmission data obtained last year (PR/NP 24, p 13) in the energy range 4 eV - 200 keV have been analysed to provide average transmissions for sample thicknesses of 10 mm - 60 mm at temperatures of 293K, 1100K and 1800K. Average transmission ratios  $T_{1100K}/T_{243K}$  and  $T_{1800K}/T_{243K}$ , corrected for the expansion of the samples and systematic errors in the background determination, were calculated in the energy range 10 eV - 200 keV. Examples of these results are shown in Fig. 3. The main features are similar to those observed in previous measurements (1,2) with uranium metal at 1100K. The maximum effect is seen in the range 500 eV - 4 keV; the position of the peak appears at lower neutron energies for lower sample thicknesses. A marked fall in the ratios is observed at 4 keV for all sample thicknesses measured. This decrease corresponds with a large fall (by possibly a factor of two) in the s-wave neutron strength function at this energy observed by Carraro and Kolar<sup>(3)</sup>.

The results at selected energies are compared with earlier calculations (PR/NP 23, p 19) in Table 1. The temperatures and sample thicknesses do not correspond exactly, but the general features of the data are reproduced, and there is quite good overall numerical agreement. In the 5 keV case it was necessary to reduce the s-wave neutron strength function to the value obtained by Carraro and Kolar<sup>(3)</sup> to obtain better agreement. As expected a larger effect is seen at the higher temperature.

The effective temperatures of the hot samples were extracted from the data by shape fitting of resonances in the resolved region using the multilevel R-matrix program REFIT<sup>(4)</sup>. The results obtained are given in Table 2, and an example of a fit shown in Fig. 4. The resonance parameters used are those obtained from an analysis of the cold sample data in conjunction with uranium solution measurements by M. C. Moxon (this report p 10). There is good agreement at the higher temperature with pyrometer measurements, and fair agreement at the lower temperature. The slight difference at 1100K is possibly due to a deposit on the quartz window of the furnace through which the sample was observed, or to a systematic error in the pyrometer (which was used near the bottom end of its usable range).

The overall good agreement between calculated and observed transmissions and transmission ratios indicates that the multilevel R-matrix theory of the nuclear

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Fig. 3. Transmission ratios for 40 mm UO $_2$  sample,  $T_{1100K}/T_{293K}$  and  $T_{1800K}/T_{293K}$ 

Temperature	Sample thick	ness (293K)	Neutron energy							
	for	for	1 keV		$5 \text{ keV}^{(a)}$		5 keV <sup>(b)</sup>		20 keV	
	calculation	experiment	theory	experiment	theory	experiment	theory	experiment	theory	experiment
Calculation 1073K	0.02425	0.0238	1.029	1.034 (0.004)	1.015	1.013 (0.008)	1.006	1.013 (0.008)	1.002	1.010 (0.008)
Experiment	0.04850	0.0474	1.045	-	1.037	-	1.017	-	1.009	-
1100K	0.09700	0.0942	1.067	1.071 (0.008)	1.075	1.034 (0.010)	1.042	1.034 (0.010)	1.032	1.028 (0.010)
	0.14550	0.1416	1.091	1.078 (0.011)	1.118	1.030 (0.021)	1.064	1.030 (0.021)	1.060	1.022 (0.021)
Calculation 2073K Experiment 1800K	0.02425 0.04850 0.09700 0.14550	0.0238 0.0474 0.0942 0.1416	1.049 1.078 1.119 1.163	1.058 (0.005) 1.062 (0.006) 1.119 (0.010) 1.222 (0.021)	1.022 1.055 1.118 1.185	1.021 (0.009) 1.025 (0.010) 1.070 (0.017) 1.123 (0.040)	1.008) 1.024 1.061 1.096	1.021 (0.009) 1.025 (0.010) 1.070 (0.017) 1.123 (0.040)	1.002 1.010 1.041 1.081	1.029 (0.009) 1.001 (0.010) 1.037 (0.016) 1.137 (0.037)

#### Comparison of calculated and observed average transmission ratios (corrected for expansion)

(b) has half the average s-wave reduced neutron width of (a) (level spacing kept constant)

Statistical errors (10) quoted for experimental values

#### <u>Table 1</u>



Fig. 4. Shape fit to the 80.8 eV resonance (10 mm UO $_2$  sample) at three temperatures

#### Table 2

#### Temperature determination of hot uranium dioxide samples by

Er (eV) (from uranium dioxide measurements)	$\Gamma_n \text{ (meV)}$	$\Gamma_{\gamma}$ (meV)	Fitted effective temperature (K) (nominal 1100K sample)	Fitted effective temperature (K) (nominal 1800K sample)
20.8711 (0.0064)	10.17 (0.10)	22.44 (0.43)	1121 (59)	1792 (169)
66.0556 (0.0177)	25.64 (0.66)	23.46 (0.87)	1130 (22)	1785 (66)
80.7616 (0.0048)	1.794 (0.058)	24 (assumed)	1121 (59)	1485 (89)
89.218 (0.046)	0.0847 (0.0051)	24 (assumed)	1127 (84)	1807 (153)
	Weighted mean (e. temperature	ffective )	1128 (19)	1788 (48)
	Absolute tempera from above	ture deduced	1125 (19)	1786 (48)
	Mean pyrometer ro	eading	1096 (9)	1792 (12)

#### resonance shape fitting (errors in parenthesis)

cross-section combined with the gas model of Doppler broadening provides a good description of the transmission of uranium dioxide at temperatures up to 1800K. A full report of this work will appear as AERE - R 8961.

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### The ${}^{239}$ Pu/ ${}^{235}$ U and ${}^{233}$ U/ ${}^{235}$ U fission cross section ratios over the energy range 0.1 to 20 MeV (G. D. James, D. B. Syme, M. C. Cooke\* and A. D. Gadd) [Relevant to request numbers: 666-7, 669, 670, 672-3; 911-5]

A measurement of the ratio of the fission cross sections of 239 Pu and 235 U has been made by neutron time-of-flight experiments on the 50m flight path of the Harwell Synchrocyclotron. Two back-to-back gas scintillation chambers were used to detect fission fragments from 20mg <sup>239</sup>Pu and 30mg <sup>235</sup>U using continuously flowing argon as the scintillation medium. Each chamber was viewed by two quartz-windowed photomultipliers. To reduce unwanted scintillations seemingly caused by fast neutrons in the quartz close to the photocathode, a four-fold coincidence was demanded between (a) pulses from each phototube discriminated at low bias, (b) a summed pulse discriminated at high bias to ensure pulse height corresponding to fission fragment energy and (c) a summed pulse discriminated at low level to provide accurate timing. The efficiency of the system was not determined and only a relative ratio of the fission cross sections has been 'obtained. This ratio has been normalised over the energy range 2 MeV - 3 MeV to the value 1.505 obtained by Carlson and Behrens<sup>(1)</sup>. The results obtained are shown in Fig. 5 together with those of the Livermore group. The data shown represent sums over 25 of the 2.5ns 1 1. 1-1. M. timing channels used in the experiment and were obtained in 150 hours of running time at a neutron pulse width of 10ns. It will be seen that the two sets of data are in good agreement from 0.1 MeV to 6 MeV. Between 6 MeV and 20 MeV the present data lie about 6% above the Livermore data and do not show the dip at 16 MeV observed at Livermore.

During the latter half of the experiment a second back-to-back gas scintillation chamber containing about 20 mg  $^{233}$ U on both sides was used to determine the  $^{233}$ U/ $^{235}$ U fission cross section ratio. Four fission yield spectra, each over 8000 timing channels, were recorded simultaneously. There was a difference of about 300mm between the flight path length for  $^{233}$ U and that for  $^{235}$ U. The results obtained, normalised to the value 1.523 obtained by Carlson and Behrens over the energy range 1 MeV - 2 MeV, are shown in Fig. 6 together with the Livermore data. At low energy the present results are slightly higher than those from Livermore whereas at high energy they are lower. This difference in shape may be due to a mismatch between the energy scales of the  $^{233}$ U and  $^{235}$ U chamber. This is unlikely and other causes for this discrepancy are being investigated.

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(1) G. W. Carlson and J. W. Behrens, Report UCRL-79577

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Fig. 5. Ratio of the fission cross sections of <sup>239</sup>Pu and <sup>235</sup>U normalised to 1.505 between 2 MeV and 3 MeV. The present data are shown by open circles and the data of Carlson and Behrens by open triangles


Fig. 6. Ratio of the fission cross section of <sup>233</sup>U to <sup>235</sup>U normalised to 1.523 between 1 MeV and 2 MeV. The present data are shown by open circles and the data of Behrens et al by crosses linked by straight line segments



#### Capture cross-section

Since the last report (PR/NP 24, p 10) corrections to the data for sample thickness effects have been carried out and the final average absorption cross-sections of  $^{241}$ Am is illustrated in Fig. 7. Multiple scattering and self-shielding corrections<sup>(1)</sup> ranged from  $^{41}$ % at 100 eV to 12% at 500 keV. The detector efficiency has been assumed constant with neutron energy since significant fluctuations in the shape of the capture gamma-ray spectrum are unlikely. The cross-section was normalized in the 1-2 keV energy range to



Fig. 7. The measured average  $^{241}{\rm Am}$  absorption cross-section in the energy range 100 eV to 500 keV

the value of 9.48 barns given by the measurement of Weston and Todd<sup>(2)</sup>. The overall uncertainty in the measurement is estimated to be  $\pm$  12% of which the largest contribution comes from background determination. The shape of the measured absorption cross-section is in reasonable agreement with the previous measurement<sup>(2)</sup> apart from a systematic departure above 10 keV which may be due to incorrect background determination or uncertainty in the shape of the incident neutron flux. Both measurements agree well with the ENDF/BIV evaluation below 10 keV but above this energy the evaluation is much lower and it appears that in this higher energy region the assumptions upon which it is based are no longer valid.

#### Fission cross-section

The fission cross-section data have now been fully processed up to a neutron energy of 10 keV and are presented as an average cross-section in Fig. 8. The sample size corrections were made using a Monte Carlo method<sup>(1)</sup> and the cross-section was normalized to that of  $^{235}$ U in the energy range 1-2 keV using a value taken from a recent status

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Fig. 8. The measured average  $^{241}$ Am fission cross-section in the energy range 50 eV to 10 keV

report<sup>(3)</sup>. Values of  $\bar{\nu}_{p}$  which were required in the analysis of the data were obtained from the publication of Manero and Konshin<sup>(4)</sup>. The cross-section was also corrected for the contribution from a <sup>239</sup>Pu impurity using the evaluated cross-section of Sowerby and Patrick<sup>(5)</sup> above 100 eV and data from the U.K. library<sup>(6)</sup> between 50 and 100 eV. The <sup>239</sup>Pu contribution was typically 50% in the 100 eV region and less than 20% at 10 keV. The principal source of error in the measured cross-section is due to uncertainties in the time dependent background. In the energy range 50 eV to 10 keV the overall uncertainty is estimated to be  $\pm 25\%$ .

The present measurements are in broad agreement with those of Bowman<sup>(7)</sup> and Shpak<sup>(8)</sup> but cast much doubt on the rather large values from the bomb shot experiments<sup>(9)</sup> above

1 keV.

- (1) M. C. Moxon, (1977) private communication
- (2) L. W. Weston and J. H. Todd, Nuclear Cross-sections and Technology, (Proc. Conf. Washington 1975) NBS Special Publication 425 (1975) 229
- (3) M. R. Bhat, U.S. Report ERDA-NDC-5/L (1976)
- (4) F. Manero and V. A. Konshin, Atomic Energy Rev. 10 (1972) 637
- (5) M. G. Sowerby, B. H. Patrick and D. S. Mather, Ann. Nucl. Sci. and Eng. 1 (1974) 409
- (6) U.K. Nuclear Data Evaluation, UKNDL DFN 269D
- (7) C. D. Bowman, M. S. Coops, G. F. Auchampaugh and S. C. Fultz, Phys. Rev. <u>137B</u> (1965) 326
- (8) D. L. Shpak, Yu. B. Ostapenko and G. N. Smirenkin, JETP Lett. 10 (1969) 175
- (9) P. A. Seeger, A. Hemmendinger and B. C. Diven, Nucl. Phys. A96 (1967) 605

#### E. <u>Capture cross-section measurements on natural iron and nickel (D. B. Gayther,</u> <u>B. W. Thomas, R. B. Thom\*, M. C. Moxon, J. B. Brisland and D. A. J. Endacott)</u> [Relevant to request numbers: 184-193, 212, 233-8, 256]

The analysis of capture yield data for iron and nickel has been taken substantially further than reported previously (PR/NP 24, p 11). Capture yields for three metallic samples (0.5, 2 and 6 mm thickness) of each material have been archived in both the coincidence and non-coincidence modes of operating the detector. The detector efficiency has been determined for each of the dominant resonances of iron and nickel below 50 keV, together with an average value for a range of resonances in the gold reference data. The techniques derived for efficiency determination are described in another contribution to this report (p 26). All yields have been normalized to the average capture crosssection of gold in the 25-35 keV energy range (assuming a value of 589 mb).

Parameters of the main resonances up to 50 keV neutron energy have been obtained by a combination of area<sup>(1)</sup> and shape<sup>(2,3)</sup> analysis programs which make corrections for finite sample effects such as multiple scattering and resonance self shielding. Average cross-sections have been determined up to 800 keV.

It is quite common to find scattering to capture ratios in excess of 10<sup>3</sup> for resonances in the structural materials and so it is necessary to make corrections to the capture yields to account for the detection of prompt scattered neutrons. This effect

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has been evaluated with a carbon scatterer and for iron and nickel the ratio of prompt neutron events to capture events is ~  $10^{-4}$ . This ratio has been used to make corrections to radiation widths obtained for the s-wave resonances where the effect is important.



Fig. 9. Fits obtained with the code FANAC<sup>(3)</sup> (solid lines) to the experimental capture yields (crosses) in the region of the 27.7 keV s-wave resonance in <sup>56</sup>Fe for three sample thicknesses of natural iron

experimental techniques and the results obtained at the present stage of analysis is given in a paper which was presented at the Specialist Meeting on Neutron Data of Structural Materials for Fast Reactors held at CBNM, Geel in December 1977 (the Proceedings of which are to be published).

A complete discussion of the

Figure 9 shows examples of shape analysis fits to a portion of the experimental data. In the figure the capture yields have been divided by the sample thickness (expressed in atoms/b); for the thinnest sample this quotient is approximately equal to the actual capture cross-section.

Future work will extend the resonance analysis and average cross-section determination to higher energies and include a similar analysis of data obtained for samples of natural chromium.

(1) M. C. Moxon, unpublished

(2) Monte Carlo program developed at Harwell by J. E. Lynn and M. C. Moxon, unpublished

(3) FANAC, F. H. Fröhner, Report KFK 2145 (1977)

#### E. <u>Capture cross-section measurements on the separated isotopes of titanium (B. Thom\*</u> D.B. Gayther, M.C. Moxon and B.W. Thomas)

Analysis has continued on capture yield data, obtained with the large liquid scintillator (PR/NP 23, pl3), for the four separated isotopes <sup>46,47,49,50</sup>Ti and a natural metal sample (principally <sup>48</sup>Ti). Detector efficiencies were calculated using the technique described in another contribution to this report (p 26). This has made it possible to derive parameters of individual resonances up to 26 keV and achieve a normalization to the gold average capture cross-section at 30 keV.

Capture yields in the energy range 9.5 - 26 keV are shown in Fig. 4. Data for  $^{50}$ Ti are not represented here as the only feature in this region is a small resonance at 17 keV. The multilevel shape analysis code FANAC,<sup>(1)</sup> which takes account of self shielding and multiple scattering effects, has been used to interpret the data. The use of such a comprehensive code is essential in this energy region where strong s-wave resonances at 16.8 keV ( $\Gamma_n = 6.4$  keV) and 22.1 keV ( $\Gamma_n = 0.442$  keV) in  $^{48}$ Ti dominate. In regions of well isolated narrow resonances, area analysis of the capture yields was used to obtain values of the thin sample "capture area",  $g\Gamma_n\Gamma_{\gamma}/(\Gamma_n + \Gamma_{\gamma})$ . Capture areas and widths of the larger resonances were corrected for the effects of prompt neutron scattering, using the measured value (10<sup>-4</sup>) of the ratio of the prompt neutron/capture detection efficiency of the scintillator (this report p 26).

The resonance parameters obtained in this analysis have been compared with an extensive set of data published recently by Allen et al<sup>(2)</sup>. Main areas of disagreement between the two data sets are in level spin and spectroscopic number assignment and also in conflicting capture areas in regions of poorly resolved resonances.

This work has been discussed in detail in a paper presented at the recent Specialists' Meeting on Neutron Data of Structural Materials for Fast Reactors (CBNM, Geel, December 1977 - to be published).

(1) F.H. Fröhner, Report KFK 2145 (1977)

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<sup>(2)</sup> B.J. Allen and A.R. de L. Musgrove, Specialists' Meeting on Neutron Data of Structural Materials for Fast Reactors, CBNM, Geel, December 1977 (to be published). Also B.J. Allen et al, Report AAEC/E402.



Fig. 4 A portion of the (capture yield/sample thickness) data for isotopically enriched samples of <sup>46,47,49</sup>Ti and a sample of natural titanium. The full curve which joins the circles is the shape fit to the data (crosses) using the code FANAC(1)

## E. Efficiency determination of a large liquid scintillator for capture cross-section measurements (B. W. Thomas and R. B. Thom\*)

In measurements of neutron capture cross-sections it is necessary to correct data for variations in detector efficiency that arise from differences in the capture gamma-ray cascade. These differences are due to fluctuations of partial radiation widths (from resonance to resonance) and mass dependent features such as binding energy and level density.

In the ideal large liquid scintillator this problem is eliminated by having sufficient detector volume to ensure that all gamma-rays are detected with 100% efficiency. Since neutron separation energies can be as large as 11 MeV, this condition, even if it could be achieved, would entail high natural background rates. In practice a compromise between background and efficiency has to be made and some dependence of efficiency on the decay mode is inevitable.

The work reported here is an integral part of a programme to determine the capture cross-sections of the reactor structural materials (Fe, Ni, Cr and Ti) using a detector with a relatively small volume of 270 1 (PR/NP 23, p 13). Since the capture gamma-ray cascades of nuclei in this mass region exhibit wide variations from resonance to resonance, it has been necessary to establish a reliable technique for deriving detector efficiencies from pulse amplitude spectra. This has added advantage that the capture cross-section of a heavy nucleus such as gold, with totally different spectra, can be used for normalization.

In the measurements themselves, both the pulse amplitude and incident neutron energy were obtained for each event. In addition a label was recorded that signified whether or not there had been a coincidence between the two halves of the detector which were optically separated by a melinex curtain. In principle, by collecting data in this manner, it should be possible to compare measured pulse amplitude spectra with those calculated from the known detector geometry and published gamma-ray spectra. The calculations will also yield values of the detector efficiency, but because gamma-ray spectra are only available for one or two resonances a semi-empirical method has been developed which is described here.

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The scheme is based on the observed pulse height distributions and calculations with a Monte Carlo code GAMOC<sup>(1)</sup>. The code predicts, for a given gamma-ray cascade, the pulse height distribution which will be observed with the detector in either its coincidence or non-coincidence mode of operation. It has been shown that spectra observed with various gamma-ray sources placed in the detector are well reproduced by the code<sup>(2)</sup>. A further test of the validity of the code has been made by comparing measured and predicted pulse height distributions for the markedly different cascades from the 1.15 keV resonance in <sup>56</sup>Fe and from averaged resonances in gold. The cascade for gold was provided by folding the known thermal neutron capture gamma-ray data with a statistical model prediction which peaked the primary gamma-ray spectrum at ~ 2 MeV, and that for <sup>56</sup>Fe was derived from published high resolution data for the resonance, coupled with the decay scheme of low lying states in <sup>57</sup>Fe determined from thermal neutron capture. The comparisons are shown in Fig. 10 where it can be seen that good agreement is obtained.

In the absence of detailed information on the gamma-ray cascade, its salient features can be related to the following properties of the observed pulse height distributions:

(a) the ratio of coincidence to non-coincidence events,  $\varepsilon_{\gamma C}/\varepsilon_{\gamma NC}$ 

(b) the shape of the distributions, characterised by a parameter F where  $\varepsilon_{\gamma C}$  and  $\varepsilon_{\gamma NC}$  are respectively the absolute efficiency of the detector above the imposed bias ("fractional efficiency"), in the coincidence and non-coincidence modes. The shape factor F, is defined in the following way. The code GAMOC is used to simulate the pulse height distribution for a single gamma-ray of energy equal to the separation energy of the compound nucleus. The distribution is not unlike that for <sup>56</sup>Fe in Fig. 10, showing a well defined peak above a valley region. A pulse height,  $E_M$ , is chosen just below the peak, and if  $E_B$  is the imposed bias level then F = (area of distribution above  $E_M$ )/(area of distribution between  $E_B$  and  $E_M$  normalized to unit height). The normalization removes the dependence of F on the separation energy.

It was established with GAMOC that, for widely differing groups of gamma-ray cascades, there exists to a good approximation a general relationship between  $\varepsilon_{\gamma NC}$  and  $F_{NC}$ . This is illustrated in Fig. 11 where each point represents a GAMOC prediction for a particular cascade. It was found that  $\varepsilon_{\gamma NC}$  could be accounted for by a constant part (36.4%) plus a component that increases rapidly as the spectrum becomes softer. The solid line is a least squares fit to the points which is given by the relation,

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Fig. 10. Measured and predicted pulse height distributions for the 1.15 keV  $^{56}$  Fe resonance and average resonances in gold

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 $\varepsilon_{\gamma NC} = 36.4 + 65 F_{NC}^{-0.88}$  (%)

The dashed lines in the figure represent the range of values required to produce 5% and 10% systematic uncertainties in  $\varepsilon_{_{\rm VNC}}$ .



Fig. 11. Monte Carlo predictions of the efficiency of the large liquid scintillator in the non-coincidence mode for widely differing gamma-ray cascades. The shape factor, F, is a measure of the "hardness" of the gamma-ray spectrum

The measurements taken in the coincidence mode are used to derive the capture crosssections, since of the two, this mode has the higher signal to background ratio and the lower sensitivity to scattered neutrons. Since a simple relation, such as the above, could not be established for  $\varepsilon_{\gamma C}$ , it is obtained by combining the derived value of  $\varepsilon_{\gamma NC}$ .

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with the observed ratio  $\epsilon_{\gamma C}/\epsilon_{\gamma NC}$ . In practice, the value of  $F_{NC}$  for a particular resonance, is obtained from the pulse height spectrum at the peak of the resonance after subtracting the pulse height spectra in the wings to allow for the effect of background. An estimated systematic uncertainty of  $\pm 0.05\epsilon_{\gamma C}$  is assigned to the values of  $\epsilon_{\gamma C}$  obtained with the scheme. In natural iron,  $\epsilon_{\gamma C}$  was found to range from 20% to 40%.

Experimental values of the efficiencies and shape factors derived in the above way were compared with GAMOC predictions for the theoretical gold and iron spectra shown in Fig. 10. The results are given in Table 1 where it can be seen that the agreement is generally good. Any differences are probably largely dependent on the gamma-ray and decay data supplied for the Monte Carlo calculations.

#### Table 1

# $\frac{\text{Comparison of experimental results and Monte Carlo predictions}}{\text{for } \frac{197}{\text{Au and the 1.15 keV resonance in } 56}{\text{Fe}}$

	197 Au - avera	age resonance	<sup>56</sup> Fe - 1.15 keV resonance		
*	Monte-Carlo experimental		Monte-Carlo	experimental	
$\epsilon_{\gamma C} / \epsilon_{\gamma NC}$	0.79	0.80	0.56	0.61	
FNC	9.0	9.0	. 15.5	16.0	
εγς	33%	36%	23%	25%	
ε <sub>γnc</sub>	42%	45%	41%	42%	

(1) R. B. Thom, Ph.D. Thesis, University of London (1978) unpublished

(2) D. B. Gayther, Report AERE - R 8082 (1975)

#### H. A random number generator for use with a small computer (M. C. Cooke\*)

Monte Carlo simulation is a powerful tool for many types of neutronics calculations. For example, it has been used to calculate the efficiency of a large liquid scintillator for capture cross section measurements (this report p 26) and to investigate by simulation observed statistical effects such as those in the fission cross sections  $^{(1,2)}$  of  $^{234}$ U and  $^{235}$ U. Such calculations can become expensive on a large computer whereas the algorithmic pseudo-random generators available on small computers often give sequences which fail tests for randomness. The availability overnight of the Honeywell DDP516 computer on the synchrocyclotron has encouraged the development of a random number generator for use on a small computer.

In this method, five digit numbers are generated by a time digitizer which is used to measure the time interval between regular clock pulses and pulses derived from the decay of a radioactive source. The digits corresponding to the tens and hundreds of these decimal numbers are concatenated in groups of three to produce the six digit numbers which are stored on magnetic tape to constitute a set of random numbers. Each tape contains 2.4 million numbers. The numbers recorded have passed stringent tests for randomness and, although from their method of generation they are truly random, they share with pseudo-random number sets the advantage of reproducibility in cases where it is necessary to repeat a calculation with the same set of random numbers. The tapes could be made available for use on any computer.

- G. D. James, J. W. T. Dabbs, J. A. Harvey, N. W. Hill and R. H. Schindler, Phys. Rev. C, <u>15</u> (1977) 2083
- (2) G. D. James, G. de Saussure and R. Perez, Trans. Am. Nucl. Soc. 17 (1973) 495

#### Resonance analysis of neutron transmission measurements on natural iron samples (M. C. Moxon and J. B. Brisland) [Relevant to request numbers: 212, 184-93]

An analysis, using the program REFIT<sup>(1)</sup>, is being carried out on transmission data obtained from measurements on samples of natural iron using the neutron time-of-flight facilities on the Harwell synchrocyclotron<sup>(2)</sup> and 45 MeV linac. The resonance parameters (for an R-matrix representation of the cross-section) obtained from the least squares fit are being used to calculate accurate corrections for self screening and multiple

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scattering for the linac measurement (this report p 24) of the neutron capture crosssection of iron.

#### (a) Synchrocyclotron data

An initial analysis of the synchrocylotron transmission data indicated the need for a renormalisation that decreased with increasing neutron energy. A background correction of several per cent in the neutron energy range  $\sim$  70 to 300 keV was also required. The renormalisation required was first thought to be due to errors in some of the initial resonance parameters but persisted after these were replaced by initial fits to all the wide resonances up to 500 keV.

After discussion with G. D. James et al<sup>(2)</sup> it was decided that the renormalisation and background corrections could be due to a component in the background that has very low transmission through the very thick resonance filters which must be used to determine the background at higher energies. This would negate the standard assumption that the attenuation of the filters for the background was exponential in form and emphasises the difficulties of accurately determining the background in the neutron energy region above 100 keV. Further experimental studies are under way.

An examination of the output parameters from REFIT indicated that, for resonances with total widths much greater than the resolution width, fits to the data gave accurate values of the resonance energy and the neutron width up to an energy of ~ 500 keV if the normalisation and background were determined for the region around the resonance at the same time as the parameters. For the narrow resonances the background could be determined from an interpolation of the values obtained from fits to the adjacent wide resonances. The resonance energy, neutron width and normalisation could then be obtained from fits to the data.

The parameters for the s-wave resonances in  ${}^{56}$ Fe up to an energy of ~ 500 keV were adjusted to give fits to the data leaving out the regions around the smaller resonances. An example is shown in Fig. 12 covering the region around the 187 keV s-wave resonance of  ${}^{56}$ Fe. Once the parameters of the s-wave resonance had been established fits were carried out to the largest of the p-wave resonances and finally to those resonances with widths narrower than the resolution function. The p-wave resonances were arbitraily split into two groups, one with spin and parity  $J^{\pi} = 1/2^{-}$  and the other with  $J^{\pi} = 3/2^{-}$ . For the wider resonances the fits indicated whether the spin was  $1/2^{-}$  or  $3/2^{-}$  but the spins could not be determined for the narrower ones.

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Fig. 12. Least squares fit to the synchrocyclotron transmission data in the energy region of the 187 keV resonance in <sup>56</sup>Fe

In refitting some of the regions around the s-wave resonance of  ${}^{56}$ Fe it was found possible to adjust the residual R-function  $R_{\infty}$  (which modifies the potential scattering cross-section) for the  ${}^{56}$ Fe s-wave component of the cross-section, but as expected there were large covariance terms between it and the normalisation of the data.

(b) Linac data

The much thinner resonance filters which could be used in the linac transmission measurements compared with those used on the synchrocyclotron meant that there was a smaller error introduced into the background by the extrapolation to zero filter thickness.

In the neutron energy region covered by the linac data (see Fig. 13) only three resonances are clearly resolved. From fits to the data between 3 eV and 3 keV it was

possible to determine the energy and neutron width of the first bound level in  ${}^{56}$ Fe. The transmission dip observed at ~ 130 eV is due to  $25 \pm 5$  ppm of cobalt and the ones at ~ 340 eV and ~ 1.0 keV are due to  $330 \pm 40$  ppm of manganese. These abundances were determined from fits using REFIT and the resonance parameters given in the latest edition of BNL 325. The small dip at ~ 10.2 keV is thought to be due to a resonance in  ${}^{58}$ Fe but no fits were carried out on it.

#### Initial Results

The resonance parameters up to ~ 220 keV determined from a least squares fit to the transmission data are given in Table 1 together with the isotopic assignment and the assumed spin and parities; the errors are only statistical. The uncertainty on the flight path length and zero time give an accuracy on the energies of ~ 1 in  $10^4$ . The uncertainty on the neutron width due to covariance terms in the fitting procedure is between one and two times the statistical error and at present is still being investigated.



Fig. 13. Least squares fit to the linac transmission data determining the neutron width and resonance energies at -3.476 keV in  ${}^{56}$ Fe, 7.782 keV in  ${}^{54}$ Fe, 3.955 keV and () 6.223 keV in  ${}^{57}$ Fe

The parameters obtained up to the present are in reasonable agreement with the values published in BNL  $325^{(3)}$  when resolution widths are taken into account.

The use of the least squares fitting program REFIT has enabled us to determine accurate values of the resonance parameters from transmission data. The ability to adjust the normalisation and background in the data was essential in analysing the synchrocyclotron data but emphasises the difficulties of measuring the background in the energy region above 100 keV and this may be a source of some of the discrepancies in previous measurements.

Transmission measurements have been carried out recently on the synchrocyclotron on 2 mm and 60 mm samples of natural iron and these data will be included in the final analysis.

- M. C. Moxon, REFIT: A least square fitting program for resonance analysis of neutron transmission data, Specialist Meeting on Neutron Data of Structural Materials for Fast Reactors, Geel (1977)
- (2) G. D. James, D. B. Syme, P. H. Bowen, A. D. Gadd and I. L. Watkins, Structural material transmission measurements on the Harwell synchrocyclotron, Specialist Meeting on Neutron Data of Structural Materials for Fast Reactors, Geel (1977)
- (3) S. F. Mughabghab and D. I. Garber, BNL-325, 3rd Edition, Vol. 1, June 1973

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Resonance parameters obtained by fitting natural iron transmission data

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Isotope	As L	sumed J <sup>π</sup>	Resonance energy (keV)	Error	Neutron width (keV)	Error
56	0	1/2+	-3.4756	0.384	0.552	0.075
56	1	1/2	1.1474	0.00045	$5.0 \times 10^{-5}$	$0.4 \times 10^{-5}$
57	0	o <sup></sup>	3.9547	0.0079	0.2126	0.016
57	0	1	6.2227	0.011	0.3683	0.019
54	0	1/2+	7.7817	0.0093	1.096	0.020
56	0	1/2+	27.8294	0.026	1.474	0.017
56	1	1/2	46.0172	0.0083	0.00594	0.00080
56	1	3/2	52.1065	0.0081	0.0144	0.00075
54	0	1/2+	52.8713	0.025	2.319	0.121
56	1	1/2	53.5311	0.013	0.00135	0.00052
56	1	1/2	55.4927	0.015	0.00084	0.00028
- 56	1	3/2	59.1846	0.0084	0.00438	0.00050
54	0	1/2+	71.8767	0.019	1.763	0.067
56	0	1)/2+	73.9599	0.0082	0.6178	0.0045
56	1	1/2	80.7432	0.0028	0.0248	0.0011
56	0	1/2+	83.5205	0.012	1.211	0.017
56	1	3/2	90.2379	0.018	0.0226	0.0014
56	1	1/2	96.2194	0.011	0.0178	0.0016
56	1	3/2	96.5117	0.018	0.0026	0.00089
54	0	1/2+	98.7106	0.077	0.5855	0.088
56	1	1/2	102.570	0.013	0.0294	0.0017
56	1	1/2	112.581	0.016	0.0161	0.0018
56	1	1/2	122.651	0.0083	0.0937	0.0024
56	1	3/2	124.054	0.023	0.0050	0.0011
56	1	1/2	124.984	0.0078	0.0258	0.0017
56	0	1/2+	129.685	0.011	0.6000	0.0088
56	0	1/2+	140.305	0.021	2.704	0.028
56	0	1/2	169.044	0.013	0.9188	0.017
56	1	1/2	173.449	0.021	0.0675	0.0054
5,6	1	3/2	179.472	0.025	0.0154	0.0018
56	1	1/2	180.904	0.014	0.0839	0.0039
56	0	1/2+	187.490	0.022	3.561	0.059
56	1	3/2	192.686	0.029	0.0282	0.0053
56	1	1/2	195.465	0.041	0.0757	0.0065
56	1	3/2	201.266	0.033	0.0367	0.0034
56	· 1	3/2	207.742	0.061	0.0131	0.0045
56	0	1/2+	220.144	0.0065	1.217	0.014
56	1	1/2	225.439	0.029	0.0722	0.0050

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H. Neutron transmission measurements on the synchrocyclotron (P. H. Bowen, A. D. Gadd, G. D. James, D. B. Syme and I. L. Watkins) [Relevant to request numbers: 165-7, 212, 184-93; 256, 233-8; 128-9, 154-5]

Since completion of modifications to the synchrocyclotron Dee structure in 1973, thirty five transmission measurements on various samples of structural materials have been carried out using the neutron time-of-flight system. An overall view of the work is presented in Table 1 which is a version of a table presented at a recent conference<sup>(1)</sup> expanded to include the five transmission measurements carried out this year. Of these five measurements, those made at the 50m detector station on iron and nickel supplement the data taken previously by extending the energy range of the measurements down to 100eV. The measurement on the 100m flight path on a 60 mm sample of iron was required to improve the resonance parameter data on narrow resonances in iron. Samples of  ${}^{50}\text{cr}_2{}^{0}{}_3$  and  ${}^{52}\text{cr}_2{}^{0}{}_3$  were recently obtained, in a joint loan to Harwell and CBNM, from ORNL. Transmission measurements at 50m for these samples are complete. Measurements at 100m will be carried out in early 1978 followed by transmission measurements listed in Table 1 is being carried out by Syme and Bowen<sup>(2)</sup> for nickel, Moxon and Brisland<sup>(3)</sup> for iron and by Thom<sup>(4)</sup> for titanium.

- G. D. James, D. B. Syme, P. H. Bowen, A. D. Gadd and I. L. Watkins, Structural Material transmission measurements on the Harwell synchrocyclotron. Proc. Specialist Meeting on Neutron Data of Structural Materials for Fast Reactors, Geel, December 1977 - to be published
- (2) D. B. Syme and P. H. Bowen, Resonance Analysis of Nickel transmission data, ibid to be published
- (3) M. C. Moxon and J. B. Brisland, Resonance analysis of transmission measurements on natural iron samples, Ibid - to be published
- (4) B. Thom private communication
- H. <u>Analysis of nickel and <sup>58</sup>Ni neutron transmission data for resonance parameters</u> (D. B. Syme, P. H. Bowen and A. T. Gadd) [Relevant to request numbers: 256, 233-8]

Nickel is an important constituent of fast reactor steels and the determination of the neutron capture cross sections of nickel isotopes has had high priority. Satisfaction of the requirements has been hampered by the lack of an adequate description of the neutron widths for resonances up to several hundred keV. Previous data were ten years old and had been analysed for resonance parameters in terms of the single-level Breit-Wigner approximation to the resonance shape. Because of the strong level-level

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Table 1

Sequence number	Sample Material	Thickness Atom/barn	Path <sup>1)</sup> (m)	Channel width (ns)	Channel Structure Reference <sup>2)</sup>	Run number	Pulses <sup>3)</sup> (10 <sup>5</sup> )
1	3 cm Fe	0.2582	100	5	ä	3499	484.7
· 2	1.2 cm Fe	0.1025	100	5	а	5498	481.9
3	3 cm Fe	0.2582	100	2.5	Ъ	7499	906.3
4	1.2 cm Fe	0.1025	.100	2.5	Ъ	7497	327.2
5	0.6 cm Ni	0.05480	100	2.5	. b ·	8497	382.9
6	1.8 cm Ni	0.1646	100	2.5	Ь	8498	657.6
7	0.9 cm Ti	0.05136	100	2.5	Ъ	9499	566.8
8	10 cm Ti	0.5706	100	2.5	Ъ	0499	1087.8
9	49TiO2	0.0379	100	2.5	c	5999	602.6
10	3.3 cm Ti	0.18654	100	2.5	c	5099	387.0
11	47TiO2	0.03924	100	2.5	c ·	5.097	518.9
1.2	46Ti02	0.03979	100	2.5	c	5197	502.2
13	3 cm Co	0.27877	100	2.5	c '	5125	346.6
14	1 cm Co	0.09327	100	2.5	с	5096	316.9
15	50Ti02	0.04694	100	2.5	с	5299	974.0
16	48 <sub>Ti</sub>	0.08239	100	2.5	· c	5298	689.8
17	3 cm Fe	0.2582	100	2.5	с	5299	302.2
18 -	46Ti02	0.3979	50	5	d ·	6199	185.5
19	47Ti02	0.03924	50	5	ď	6198	235.7
20	<sup>48</sup> Ti	0.08239	100	2.5	с	6294	484.0
21	50 <sub>Ti02</sub>	0.04694	100	2.5	с	6298	938.8
22	50TiO2	0.04694	50	5	d	6297	187.3
23	48 <sub>Ti</sub>	0.08239	50	5	· d	6296	275.2
24	49Ti	0.0379	50	5	d	6295	265.6
25	58 <sub>Ni</sub>	0.08458	50	5	d	6799	237.2
26	58 <sub>Ni</sub>	0.08458	100	2.5	с.	6898	850.2
27	1.2 cm Fe	0.1025	100	2.5	c	6896	511.0
28	58 <sub>Ni</sub>	0.08458	100	2.5	c.	6895	526.6
29	0.6 cm Ti	0.03366	50	5	d	6698	82.4
30	1.8 cm Ti	0.10197	50	5	d	6997	114.7
31	6 cm Fe	0.51513	100	2.5	c	7099	321.1
32	0.06 cm Ni	0.00572	50	5	d	7199	155.1
· 33 ·	0.2 cm Fe	0.01698	50	5	d	7198	282.8
34	52Cr203	0.1189	50	5	d	7197	399.4
35	50Cr203	0.07249	50	5	d ,	7299	
1	1			1 ,	1	1	

List of transmission measurements made from 1974 to 1976

1) Runs 1 and 2 and all 50m runs are taken with the  ${}^{6}$ Li-glass detector. All other runs are taken with the NE110 detector

2) The channel structure implies a doubling of channel width at the following channels:-

(a) 16000, 26000, 28000

(b) 26000, 28000

(c) None

(d) 13000, 23000

3) The number of pulses given is for the sample in run only. The number of pulses in the sample out run is adjusted to roughly equal the number of counts in the peak of the two spectra

Experiments at 100m are performed at 800 pps. Those at 50m are performed at 400 pps

interference of l = 0 resonances in the structural materials, the single-level approximation does not give a good description and a multi-level treatment is to be preferred.

Several recent data on transmission of neutrons through natural nickel (PR/NP 22, p16) and <sup>58</sup>Ni (PR/NP 24, p14) have been under analysis here. It would be best to treat resonance states of all angular momenta explicitly by multi-level R-matrix analysis. This is however slow and expensive in computer time and our procedure has been to analyse:

- 1. the narrow (orbital angular momentum,  $\ell > 0$ ) resonances by single level area analysis up to an energy limited by resonance overlap, and
- 2. the broad and overlapping s-wave (l = 0) structure by a multi-level R-matrix code up to an energy where it is necessary to treat the l > 0 resonances explicitly.

These two stages have been concluded and their results described in the recent Geel meeting<sup>(1)</sup>. Some of the main features are described in section (1) and (2). We have combined the information from stages (1) and (2) and are proceeding to higher energies with a full multi-level analysis.

 Single level area analysis of l > 0 resonances in nickel and <sup>58</sup>Ni transmission data (P. H. Bowen and D. B. Syme)

We have completed the area analysis of narrow (l > 0) resonances in natural nickel described in PR/NP 24, p12 and the resonance parameter set obtained for <sup>58</sup>Ni has been extended to include 97 resonances by similar analysis of recent synchrocyclotron transmission data on <sup>58</sup>Ni (PR/NP 24, p14). These were presented at the Geel meeting<sup>(1)</sup>. Comparison with the few previous data is fairly good if one allows for our better neutron energy resolution. For example, at 215 and 247.5 keV we have now resolved doublets (see Table 1); the sum of the two new widths agrees with the older width in each case. In preparation for the next stage of analysis, a comparison has been made of the fits to three well-resolved narrow resonances in <sup>58</sup>Ni by both the area analysis<sup>(2)</sup> and Rmatrix<sup>(3)</sup> codes. An example of the excellent agreement in fit and extracted neutron widths is shown in Fig. 14.

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Comparison of l > 0 resonance parameters

in <sup>58</sup>Ni

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BNL 325		Present Data $\frac{\Delta E}{E} \sim 10^{-3}$		
E(keV)	Γ <sub>n</sub> (eV)	E(keV)	Γ <sub>n</sub> (eV)	
$147.5 \pm 0.8$ $183.5 \pm 1.1$	175 250	148.73 184.53	$\begin{array}{r} 136.5 \pm 3.5 \\ 131 \pm 9 \end{array}$	
215.0 + 1.5	260	216.52 217.90	200 <u>+</u> 14.5 34.5 <u>+</u> 9.5	
247.5 <u>+</u> 1.8	360	249.39 250.06	268 <u>+</u> 20 68 <u>+</u> 16	
286.5 <u>+</u> 2.0	215	289.30	210 <u>+</u> 20	

Fig. 14. Comparison of fits and resonance parameters for the 82.8 keV p-wave resonance in  $5^{8}$ Ni using the Atta-Harvey area analysis code<sup>(2)</sup> and the R-matrix multi-level code of Moxon<sup>(3)</sup>. ( $5^{8}$ Ni data)

# (2) Single channel multi-level R-matrix analysis of l > 0 resonances in <sup>58</sup>Ni and natural nickel transmission data (A. T. Gadd and D. B. Syme)

We have analysed transmission data to obtain s-wave resonance parameters using a single channel multi-level R-matrix code up to the energies where  $\ell > 0$  resonances require to be considered explicitly in each data set. Initial lists of resonance parameters were presented at the Geel meeting for <sup>58</sup>Ni (15-300 keV) and <sup>60</sup>Ni (12-200 keV). Most of the present widths are smaller than those in BNL 325 and some discrepancies in the energy scale and spin assignments of the earlier data have been revealed. For example the 83.1 keV resonance in <sup>58</sup>Ni believed to have spin and parity  $J^{\pi} = \frac{1}{2}^{+}$ ,  $\ell = 0$  is clearly observed as a  $\ell > 0$  doublet in our data (one component was shown in Fig. 14) and the 36.1 keV resonance in <sup>58</sup>Ni has now been clearly assigned  $\ell = 0$ , not  $\ell > 0$ . The fit to the 36.1 keV resonance is shown in Fig. 15.

The set of s-wave parameters will not be final until our present analysis has been extended to much higher energies, because of the strong resonance-resonance interference





<sup>58</sup>Ni SAMPLE FITTED FOUR RESONANCES E = 108 · 15 keV, GN = 1 · 072 keV E = 123 · 38 keV, GN = 0 · 435 keV  $E = 137 \cdot 32 \text{ keV}, \text{ GN} = 2.617 \text{ keV}$ E = 139 91 keV, GN = 2.867 keV 1.0 Transmission 0.2



Fig. 16. Fit by an R-matrix multi-level code  $\binom{(3)}{a}$  to four 58 Ni s-wave levels in a 58Ni sample

seen in the typical fit in Fig. 16.

(3) Computer code for resonance parameter distributions (A. T. Gadd and D. B. Syme)

The lists of resonance parameters (e.g. neutron widths at each energy) obtained from area and shape analysis are read by a new computer code which converts each list to the various conventional forms used to determine average resonance parameters and show statistical distributions.

The widths are plotted against energy then converted to reduced widths. From plots against energy of cumulative resonance number and cumulative reduced width the mean spacing and strength function are found by least squares fits. It is a feature of the program that this may be done for any specified energy range, so that the possible variation of these average parameters in different regions (due to the intermediate structure effects expected in the structural materials) may be quantified. The number distribution of level spacings (nearest and next nearest neighbour) and reduced widths are also calculated and plotted. We intend to add comparisons with the corresponding theoretical distributions in each case.

- (1) D. B. Syme, P. H. Bowen and A. T. Gadd, presented at the Specialist Meeting on Neutron Data of Structural Materials for Fast Reactors, CBNM, Geel, December 1977
- (2) S. E. Atta and J. A. Harvey "Numerical Analysis of Neutron Resonances", Report ORNL 3205 (1961)
- (3) M. C. Moxon "REFIT", A least square fitting program for resonance analysis of neutron transmission data", presented at the Geel meeting (as in ref. (1))

#### H. Neutron energy standards (G. D. James)

The work of an INDC ad-hoc sub-group on neutron energy standards was completed by the selection of 41 narrow resonances in the energy range 0.1 eV to 12.1 MeV which should prove suitable for the intercomparison of neutron energy scales. The sub-group consisted of J. Boldeman, F. Corvi, J. A. Harvey, G. D. James (Co-ordinator), J. Lachkar, A. B. Smith and F. Voss. The resonances selected, identified by their nominal energies, are given in table 1. Comparison of energy determinations<sup>(1)</sup> for a few resonances show that at best resonance energies can be quoted to an accuracy of one in 10,000. Not all the energies listed in table 1 are known to the highest energies attainable.

 G. D. James, Neutron energy standards, Neutron Standards and Applications, NBS Special Publication 493 (1977) 319

	<del></del>		
Energy Range (eV)	Isotope	Nominal Energy (eV)	Order*
0.1 - 1	<sup>191</sup> Ir	0.6551 <u>+</u> 0.0014	1
1 - 10	238 <sub>U</sub>	6.672	1
$10 - 10^2$	238 238 <sup>U</sup>	10.236	5
	238 <sup>U</sup>	20.864	4
	238 <sup>0</sup>	66.015	2
	238 <sub>U</sub>	80.729	1
$10^2 - 10^3$	238 238 <sup>U</sup>	145.617 + 0.033	7
	238 <sup>U</sup>		8
	238 <sup>0</sup>	397.58	4
	238- 238 <sup>0</sup>	463.18 <u>+</u> 0.24	3
	238 <sup>U</sup>	619.95	2
	238 <mark>0</mark>	$708.22 \pm 0.20$ 905.03	6
$10^3 - 10^4$	238	1419.88 + 0.32	7
	238 <sup>-</sup> 238 <sup>0</sup>	1473.8	. 5
	238 <sup>U</sup>	$2489.47 \pm 0.5$	3
	206 <sup>0</sup> Pb	$3360 \pm 10$	8
	238 <sup>1</sup>	3458.1	6
	238 <sup>U</sup>	4512.0	2
	27.1	5650.6	1
(1 37)		(1 - 11)	9
(KeV)	32_	(kev)	
$10 - 10^{-1}$	23 <sub>N2</sub>	$\begin{array}{c} 30.378 \pm 0.006 \\ 53.101 \pm 0.027 \end{array}$	3
		67.73 + 0.02	5
	206 56 <sup>Pb</sup>	71.191 + 0.018	4
	32 <sup>Fe</sup>	$90.134 \pm 0.016$	2
3	32	97.512 + 0.028	
$10 - 10^{-1}$	56 <sub>F</sub>	$112.186 \pm 0.033$	
	32 <sup>r</sup> e S	412.3	1
,	<sup>32</sup> s	818.7	6
$10^3 - 10^4$	<sup>16</sup> 0	1651 + 2	5.
	$^{12}_{12C}$	2078	4
	1 <sup>2</sup> C	2818 + 4	3
	120	3211.1 + 1.5	
1.45	12		<u> </u>
10 - 10	С	$12100 \pm 100$	1

Narrow resonances suitable for use as energy standards

Table 1

\*Order of  $(\Gamma + \Delta)/E$  within each energy range defined above. The smallest number corresponds to the narrowest resonance in each range. D. <u>Measurement of the fast fission neutron spectrum of</u> <sup>235</sup>U at 0.1 MeV incident neutron <u>energy (J. M. Adams, B. Trostell\* and L. Eriksson\*)</u> [Relevant to request numbers: 767, 769]

The analyses of the experimental data (PR/NP24, p20) have not yet been completed, but the results to date confirm that a Watt formalism provides a better description of the shape of the fission neutron spectrum than a Maxwell formalism (PR/NP 23, p 21). Currently the analysis is concerned with the correction of the data for the finite size of the <sup>235</sup>U sample used for the experiment. In this connection a comparison is being attempted between an analytical and a Monte Carlo technique, in order to investigate the overall validity of the much simpler first order analytical approach. For this purpose, the computer program MAGGIE<sup>(1)</sup> is being modified to expand the non-elastic scattering aspects more explicitly in order to obtain the appropriate multiple scattering correction factors as a function of fission neutron energy.

 J. B. Parker, J. H. Towle, D. Sams and P. G. Jones, Nucl. Instr. and Methods <u>14</u> (1961) 1 and J. B. Parker, J. H. Towle, D. Sams, W. B. Gilboy, A. D. Purnell and H. J. Stevens, Nucl. Instr. and Methods <u>30</u> (1964) 77

#### D. Detector calibration for neutron flux measurements (C. A. Uttley, J. A. Cookson, M. Swinhoe\*\* and C. Wise\*\*)

The measurement of the absolute efficiency of a 5 cm diameter by 3.8 cm thick NE213 liquid scintillator to neutrons over the energy range 1.5 to 25 MeV has been completed. The associated particle method was used as discussed in PR/NP 23, p19 employing the  $T(p,n)^{3}$ He,  $D(d,n)^{3}$ He and  $T(d,n)^{4}$ He reactions. An n- $\gamma$  discrimination system was employed so that the primary neutron flux from various neutron source reactions could be determined for cross section measurements in the MeV region of neutron energies. An advantage of the associated particle method is that the pulse amplitude distribution in the scintillator for monoenergetic neutrons is measured over the energy range of the calibration and enables the spectrum of secondary neutrons from source reactions to be determined in addition to the primary flux. The efficiency for two proton bias values is shown in Fig. 17, along with the efficiency predicted by the Stanton Monte Carlo code <sup>(1)</sup> which includes some recent modifications by McNaughton et al.

\*Neutron Physics Laboratory, AB Atomenergi, Studsvik, Sweden \*\*Physics Department, University of Birmingham



Fig. 17. Absolute neutron detection efficiency of NE213 liquid scintillator at 1.4 and 4 MeV proton biases. Curves are predicted efficiencies from the modified Stanton Monte Carlo  $code^{(1,2)}$ 

- (1) N. R. Stanton, COO-1545 92 (1971)
- (2) M. W. McNaughton, N. S. P. King, F. P. Brady and J. L. Ullmann, Nucl. Instr. and Methods 129 (1975) 241
- D. The  $\frac{7}{1240-4}$  [Relevant to request numbers: 1240-4]

The cross section for this reaction is being measured from its threshold near 2.7 MeV to 14 MeV by irradiating a lithium compound enriched to 99.97% in lithium-7 with monoenergetic neutrons and measuring the tritium produced by  $\beta$  counting. The lithium samples are 1 gm compacted discs of either  $\text{Li}_2\text{CO}_3$  or LiOH.H<sub>2</sub>O. The former require a camphor binder which is subsequently evaporated prior to irradiation. The lithium hydroxide monohydrate has recently been obtained with the required high enrichment and requires no binder. The tritium produced in a sample following irradiation is measured by dissolving it in acid and adding liquid scintillant<sup>(1)</sup>, the counting efficiency having been previously determined for the solution from quench curves measured using a standard solution of tritiated water obtained from RCC Amersham. At present both hydroxide and carbonate samples are being used in the irradiations because the labile fraction of tritium stored in the former compound is known to be 99.7% but has not been measured for the carbonate. (The labile fraction is the tritium in the form T<sup>+</sup> which goes readily into solution in the scintillant mixture.) Samples of natural lithium carbonate and hydroxide have been irradiated in the

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GLEEP reactor in order to determine the labile fraction in the carbonate approximately. This indicates the labile fractions of the two compounds are similar. Samples of  $\text{Li}_2\text{CO}_3$ and  $\text{LiOH}.\text{H}_2\text{O}$  enriched to 98% in lithium-6 have been prepared for a further reactor irradiation to determine the labile fraction in the carbonate more precisely and also to measure the thermal <sup>6</sup>Li(n,  $\alpha$ )T cross section as a check on the reliability of the technique.

(1) R. Dierckx, Nucl. Instr. and Methods 107 (1973) 397

D.  $(n,\alpha)$  cross sections (J. A. Cookson and C. Wise\*) [Relevant to request numbers: 1369-71, 1394-96, 1417-19]



Fig. 18. A schematic view of the counter telescope built for  $(n, \alpha)$  measurements. The tessource of neutrons will be about 5 cm from the target and the detector can be rotated about a vertical axis through the target for measuring angular distributions of  $\alpha$ -particles

An experiment to measure the  $(n,\alpha)$  cross sections of Fe, Ni and Cr at energies of interest for fusion reactors has been started. The initial measurements will use 14 MeV neutrons from the Cockcroft-Walton accelerator and natural targets of Fe and Ni.

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A special detector for this work has been constructed and is shown in Fig. 18. It consists of a telescope of four proportional counters and a CsI scintillator. Three of the proportional counters are used in coincidence with the CsI while the fourth is used in anticoincidence. Initially the counter gas will be hydrogen with a little methane, and alpha particles will be identified by measurements of  $\Delta E$  in a proportional counter and E in the CsI. The 14 MeV neutron flux will be monitored by using a surface barrier detector to record alphas from the T(d,n) $\alpha$  reaction in a solid target.

\*Physics Department, University of Birmingham

D.  $(\alpha, n)$  yields from light elements (D. West and A. C. Sherwood) [Relevant to request numbers: 19, 53, 59, 1525, 1526, 1528]

A large fraction of the preliminary work necessary before actually taking data has now been completed. The following items have required effort in approximately equal amounts.

- (1) Setting up nine individual neutron counting channels and establishing arrangements for intercomparing the channels and for calibrating the whole assembly.
- (2) Commissioning and testing the Moderator Assembly.
- (3) Final design and testing of target arrangements to ensure Faraday cup reliability without interfering with the neutron counting.
- (4) Beam energy measurements using Rutherford and elastic nuclear scattering of α particles from thin gold foils mounted on carbon.
- (5) Testing and modifying a Tandem beam line to enable both energy and neutron yield measurements to be carried out without readjusting the accelerator.
- (6) Light element target testing for the presence of neutron producing impurities.
- (7) Data handling.

It is not proposed to discuss all these topics further but to confine attention to the commissioning of the moderator assembly, beam energy measurement and light element target testing.

(1) Moderator assembly

Figure 19 shows a photograph of the moderator assembly. It was received from workshops at the end of August and consists of a lm right cylindrical block of polythene made up of slabs 100 mm thick. Nine <sup>3</sup>He/Argon-methane filled counters 19 mm in diameter and Im long are placed in holes parallel to the axis, each at a different radius, to sample the thermal neutron distribution generated from a neutron source placed at the centre. A central core (removeable) accommodates the beam pipe and the innermost three counters. Surrounding the moderator is a borated resin shield. The whole is mounted on runners to allow for frequent withdrawal from the beam pipe for replacing targets or the insertion of quartz windows for visual inspection during beam refocusing which is necessary at each change of beam energy.

A large number of neutron counting tests have been carried out. The most significant concern the required independence of efficiency to neutron energy in the range 0-12.5 MeV.

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Fig. 19. Shielded neutron moderator assembly for measuring neutron yields from  $(\alpha,n)$  reactions at the Tandem generator

Four neutron sources of different energy spectra, previously calibrated at NPL, were compared. The results for 8 and 9 counting channels (an additional channel at 57.5 mm radius) are shown in Table 1. This gives  $\Delta_{\text{Here}} - \text{N.P.L.}$  the percentage difference in the ratio of strength of a particular source to that of Am/Be measured here and at NPL. The average energy of Am/Be neutrons is 4.5 MeV and the maximum is 10 MeV.

Tabl	le 1

Source	Average Energy (MeV)	Maximum Energy (MeV)	∆ <sub>Here - N</sub> 8 Channel	.P.L.(%) 9 Channel
<sup>241</sup> Am/B	2.8	5.0	+ 0.245 <u>+</u> 0.67	+ 0.54 <u>+</u> 0.69
<sup>241</sup> Am/F	1.3	2.5	+ 0.29 <u>+</u> 0.63	+ 0.05 + 0.62
<sup>241</sup> Am/Li	0.5	1.4	- 1.66 <u>+</u> 0.96	- 2.12 <u>+</u> 0.68

Apart from the <sup>241</sup>Am/Li source, agreement with the NPL ratios is obtained within 0.5%. The discrepancy in the case of <sup>241</sup>Am/Li was due to the low energy of the neutrons; an additional counting channel near to the axis was used. A 2% discrepancy with the NPL value persists for this source and is being investigated further.

Tests of the mutual effect of the holes containing counters have shown (by plugging all holes except one in turn) that this causes a negligible difference  $(0.07 \pm 0.18\%)$  in measured source strength.

The background counting rate in quiet conditions amounts to 0.2 to 0.3 counts per minute in all channels and is equivalent to that produced by a neutron source equal to 40 neutrons/s at the centre. Compared with the strength of the calibrated Am/Be source, which is well suited to the counting system giving 30,000 c.p.m. in the innermost channel, this neutron background amounts to a fraction 1.5 x  $10^{-4}$  of the source strength.

#### (2) Tandem beam energy measurement

Rutherford scattering from a thin gold foil at approximately  $45^{\circ}$  into a Si detector is used in conjunction with elastic scattering from the carbon backing to determine both the primary beam energy and the exact mean scattering angle. Energy calibration is performed with <sup>244</sup>Cm alphas of 5.805 MeV energy. Four measurements extending over the past year have been analysed so far. For a nominal beam energy of 7.500 MeV, reset for each measurement, and usually taken on different days, the energies determined have a mean value of 7.479 MeV with a population standard deviation of 9 keV. The accuracy of an individual measurement is ~ 1 keV. The 9 keV standard deviation is indisputable and typically would cause 2% change in thick target ( $\alpha$ ,n) yield if not allowed for. Part of the difference between 7.500 MeV and the observed 7.479 MeV may be due to a dead layer on the Si detector but only 5 keV energy change is expected from this case. Steps are in hand to measure the dead layer effect by inclining the counter.

(3) Target testing for the presence of neutron producing impurities

Observations by Lees<sup>(1)</sup> of Be impurities in some light element ( $\alpha$ ,n) sources lead us to fear that similar contamination might occur in the light element targets to be used for ( $\alpha$ ,n) yields. Accordingly, we examined the  $\gamma$ -rays emitted at 90° to the beam direction from a variety of light element targets using a 90 cc Ge(Li) detector. Thick targets of Be, B, C, F, Al and Mg were examined at incident energies of 4, 5.5 and 7.5 MeV in most cases for a given incident quantity of charge. Measurements of neutron production in a 50 cm diameter block of polythene with a pair of counters then enabled the neutron and  $\gamma$ -ray yields to be related. All the targets tested gave readily identifiable  $\gamma$ -ray lines from the product nucleus following ( $\alpha$ ,n) reactions as well as  $\gamma$ -rays following ( $\alpha$ ,p) or ( $\alpha$ , $\alpha$ ') reactions in many cases. Usually there were several lines from a given nuclide except for <sup>10</sup>B which gives none and <sup>9</sup>Be which gives only one. Choosing the most prolific  $\gamma$ -ray as an indicator for each element (A) one can estimate from the background level at that energy in the spectrum from any other target (B) the level of impurity in terms of percentage of neutrons which can be detected by looking for A's  $\gamma$ -rays in B's spectrum.

These limiting percentages are given in Table 2 for the light elements studied except Mg which was not measured at the bombarding energy of 7.5 MeV. The  $\gamma$ -rays for which the limits are calculated are given in each case. Levels of neutron impurity in the few percent region are evidently detectable in all these light element targets and the technique will be useful for quality control on the targets to be used in the yield measurements. Since no impurities were in fact found in any of the targets (except carbon in the PTFE Fluorine target) the table also indicates the neutron impurity limits in the targets tested.

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#### Limiting percentages of neutrons arising from specified impurities which

could be detected in 45 minutes counting with a 90 cc Ge(Li) detector

				/	
IMPURITY MAIN	Ве	В	С	F	Al .
Be		0.7	0.1	0.08	0.29
B	1.0		0.07	0.11	0.45
С	3.4	0.9		0.11	0.35
· F	1.2	0.8	0.09		0.9*
A1	1.8	2.2	0.17	0.22	
Energy and origin of γ-radiation	4.439 MeV $9_{Be} (\alpha, n)^{12}C$ Exothermic	1.632 MeV 11 <sub>B</sub> (α,n)14 <sub>N</sub> Threshold 5.164 MeV	6.130 MeV 13 <sub>C</sub> (α,n)160 Threshold 5.12 MeV	0.583 MeV $19_{F} (\alpha, n) 22_{Na}$ Threshold 3.067 MeV 0.891 MeV Threshold	1.454 MeV 27A1 (α,n)30p Threshold 3.843 MeV *1.973 MeV Threshold 5.295 MeV

for bombarding energy of 7.5 MeV

(1) E. Lees, this progress report, p 53

### E. Neutron production from $(\alpha, n)$ reactions in <sup>241</sup>AmO<sub>2</sub> (E. W. Lees)

An improvement in the knowledge of the yield of neutrons resulting from  $(\alpha,n)$ reactions in oxide fuel is desirable from operational and environmental considerations in the nuclear fuel cycle. Measurements to date have been obtained using monoenergetic  $\alpha$ -particles incident upon a thin target of the light element, whereas the quantity of interest to the nuclear power programme is the thick target yield. Accordingly, such quantities have been calculated from the known  $(\alpha,n)$  cross-sections and the stopping powers of  $\alpha$ -particles in the material in question<sup>(1,2)</sup>, and the aim of the present study is to compare theoretical and experimental estimates.

The neutron yield from ( $\alpha$ ,n) reactions on  $^{17}$ 0 and  $^{18}$ 0 in a 12 g  $^{241}$ AmO<sub>2</sub> sample was

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measured using the large BF<sub>3</sub> detector assembly built for photofission and photoneutron studies (PR/NP 24, p39). The initial result was more than a factor of two in excess of the figure expected from theoretical considerations. This lead to an investigation of possible impurities present:

- (a) by measuring the neutron coincidence rate, it has been established that more than 98% of the source emission is due to random  $(\alpha,n)$  production, i.e. the discrepancy is not due to the presence of a spontaneous fissioning impurity:
- (b) by measuring the gamma-ray spectra to be expected from commercial  $(\alpha,n)$  sources  $(^{241}Am/Li$ , Be, B and F), and comparing with the spectra from the 12 g AmO<sub>2</sub> sample (Figs. 20 and 21), it has been established that there is clear evidence for an F impurity.



Fig. 20. The gamma-ray spectrum from the 12 g  $^{241}$  AmO<sub>g</sub> sample between 0.8 and 2.65 MeV. Note the presence of both F, 170 and <sup>18</sup>0 reaction gamma-rays. The labelling notation is: F.E. = full energy peak, S.E. = single escape peak, D.E. = double escape peak, C.E. = Compton edge. All energies are in MeV



Fig. 21. The gamma-ray spectrum from the 12 g <sup>241</sup>AmO<sub>2</sub> sample between 2.65 and 4.55 MeV. See Fig. 20 for labelling notation

The neutron strength of the commercial Am/F sources had been previously calibrated, and thus it was possible to relate the gamma-ray and neutron intensities. After correcting for the different gamma-ray attenuations in the sample and the Am/F source containers, the result for the neutron production from  $^{241}$ AmO<sub>2</sub> is  $(2.78 \pm 0.41) \times 10^3$  n/s/g  $^{241}$ Am, which is consistent with the theoretical estimate of 2.0 x  $10^3 \pm 30\%$  n/s/g  $^{241}$ Am. Note that the presence of a small F impurity (~ few parts per thousand by weight) accounts for approximately half the neutron activity of the 12 g AmO<sub>2</sub> sample and indicates the importance of sample purity in such measurements. This work has been submitted for publication.

- (1) J. E. Lynn and M. G. Sowerby, Neutron emission in fast reactor fuels, AERE Harwell (unpublished)
- (2) H. Liskien and A. Paulsen, Atomkernenergie 30 (1977) 59

Ε.

Gamma-ray spectra from commercial  $(\alpha,n)$  sources and their use in detecting source impurities (E. W. Lees and D. Lindley\*)

This work developed as an offshoot from the studies of neutron production from  $(\alpha,n)$  reactions in <sup>241</sup>AmO<sub>2</sub> (this report p 50). The aim was to determine the characteristic spectra of gamma-rays emitted by light elements following  $\alpha$ -reactions using commercially available neutron sources consisting of <sup>241</sup>Am with either F, B, Be or Li and, if possible, to use these spectra to estimate the contribution of these elements to the total neutron output of the <sup>241</sup>AmO<sub>2</sub> sample.

The majority of the spectra were taken with an 80 cm<sup>3</sup> co-axial Ge(Li) detector and an example from an Am/B commercial source is shown in Fig. 22. In addition to the 3.684 MeV and 3.854 MeV gamma-transitions arising from  ${}^{10}B(\alpha,p){}^{13}C*$  reactions, notice the presence of the  ${}^{9}Be(\alpha,n){}^{12}C*$  line at 4.439 MeV. All the commercial ( $\alpha,n$ ) sources





\*Downing College, University of Cambridge

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contained Be as an impurity and the estimates of the impurity contributions to the neutron  $\frac{1}{1}$  production were achieved by comparing the gamma-ray spectra of calibrated and unknown samples. The total neutron outputs of the ( $\alpha$ ,n) sources were calibrated by the Radiation Physics Division of the National Physical Laboratory. After correcting for the differing attenuations of the gamma-rays in the source containers, the final results for the Be impurities to the total neutron output are shown in Table 1.

 $\pm 1$ 

	Summ	ary of measured im	purities in (α,n) s	sources
·		r		······
: *.	Source Type	Serial No.	Total Activity (10 <sup>4</sup> n/s)	% due to Be
	Am/B	AMN-1000-7584B AMN-20-7427B	$\begin{array}{rrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrr$	$\begin{array}{rrrr} 0.26 & \pm & 0.05 \\ 2.71 & \pm & 0.35 \end{array}$
i	Am/F	AMN-1000-7581F AMN-1000-7582F AMN-100-5828F	$\begin{array}{rrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrr$	$\begin{array}{rrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrr$
	Am/Li	AMN-100-5831Li	0.7054 <u>+</u> 0.3%	5.42 <u>+</u> 0.84

Tab]	le l
	-

# Since the neutrons produced by ${}^{9}Be(\alpha,n)$ reactions have a maximum energy of 11 MeV they have a higher energy than those produced from the major reaction in the source, e.g. Li - 1.5 MeV. Consequently, this may have a disturbing effect in investigations where the neutron energy spectrum is of importance.

The work has also demonstrated the usefulness of Ge(Li) detectors to produce estimates of low levels of light elements present as impurities in  $\alpha$ -active samples, e.g. the Be content of the sources described is typically a few parts per thousand by weight, yet is easily and precisely detected in the gamma-ray spectra. The three most prolific ( $\alpha$ ,n) emitters of the light elements are easily identified because of the high energies of the gamma-rays (4.439 MeV - <sup>9</sup>Be, 3.684 MeV - <sup>10</sup>B, 3.182 MeV - <sup>19</sup>F) which reduce problems of attenuation in the sample and also place the observed lines much higher in energy than those gamma-rays resulting from fission products or actinide layers. In addition, the Doppler-broadened shape of the lines resulting from ( $\alpha$ ,n) reactions will further facilitate identification.

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The work is to be published as AERE - R 8891.

### Fast neutron activation of A1, Si and F (B. W. Hooton and A. Talbot)

The fast neutron activation of Al, Si and F is being investigated because of its possible use as a method for counting fast neutrons in the presence of low energy neutrons and gamma-rays. The interest lies in the neutron interrogation of fissile materials using a primary neutron source of energy below the  $^{238}$ U fission threshold. In such a case a measurement of delayed neutrons, when the interrogating source has been removed, can only utilise about 1% of the neutrons generated. A measurement of prompt neutrons is therefore extremely advantageous but necessitates a measurement in the presence of the intense interrogating source. The use of a threshold reaction with a Q value greater than the energy of the interrogation source might be of value if a sufficiently sensitive method could be found. In general terms, reactions of the type (n,p) (n, $\alpha$ ) have low cross sections, and comparison with a fast fission counter would tend to suggest that activation would have no particular benefit. However, the detection sensitivity is a product of cross section and the amount of material which can be utilised, which in the case of activation by (n,p) and (n, $\alpha$ ) reactions can amount to several hundred grams.

Samples of A1, Si and LiF have been activated using neutrons from a  $^{252}$ Cf source of 2 x 10<sup>6</sup> neutrons/s. The samples were irradiated, transferred to a Ge(Li) detector and counted over several counting cycles. The cycles were optimised by computing a figure of merit for various irradiation, counting and transfer times. Two figures of merit are relevant in practice; the signal obtained per unit time overall, and the signal relative to the background.

Reaction	Q	Activity Half-Life	E <sub>Y</sub> (MeV)	Fission Spectrum Activation Cross Section (mb)
<sup>28</sup> Si(n,p) <sup>28</sup> A1	-3.85	2.3m	1.78	7.25
<sup>27</sup> Al(n,p) <sup>27</sup> Mg	-1.83	9.46m	0.84 1.01	3.25
$19_{F(n,p)}$	-4.03	29s	0.197	0.85
$19_{F(n,\alpha)}^{16}$	-1.51	7.2s	6.13	11.7

#### Table I

Neutron activation data

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by the <sup>28</sup>Si(n,p)<sup>28</sup>Al reaction. Three cycles of 3m irradiation, 0.25m transfer and 3m counting

All the reactions given in Table ! were observed with relative intensity in approximate agreement with the activation cross sections. The spectrum for activation of  ${}^{28}$ Si(n,p)  ${}^{28}$ Al is shown in Fig. 23, for a sample consisting of 1685 g of amorphous silicon in ten polyethelene bottles strapped to the outside of a 10 cm diameter beaker. In the case of the  ${}^{28}$ Si(n,p)  ${}^{28}$ Al, reaction. where the signal to background was high, measurements were also made using a NaI detector. An activity in the samples could arise from thermal neutron capture or long term build up of other activities. In the present experiments the thermal neutron flux was negligible and no build up of long lived activity was observed following several days continuous irradiation. No

competing reactions are expected to interfere with the measurements when lifetimes and energy selection in the detector are taken into account.

If this technique of fast neutron measurement is applied to neutron interrogation of fissile material then operational constraints may decide which reaction, by virtue of its half-life, would be most convenient. An operational system might involve a transfer of activated material by means of a continuously pumped liquid, which could be rapidly removed to a remote, and well shielded, counting station. A system of this type is being constructed for tests on  $UO_2$  samples enriched to 3% in  $^{235}U$ . In the first experiment the circulating liquid will be a fluocarbon compound which should yield better data on the  $^{19}F(n,p)$  and  $^{19}F(n,\alpha)$  reactions than was obtained by manual transfer. The system will use low energy neutrons from the  $^{7}Li(p,n)$  reaction and should provide a valuable insight into the viability of the technique.

### The design of systems for determination of plutonium by passive neutron counting (B. W. Hooton)

This work concerns calculations which have been carried out with a specific objective in mind, namely, the design of systems for the determination of plutonium in the raffinate

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streams of a nuclear fuel reprocessing plant. The results can however be applied to the design of almost any system based on a thermal neutron measuring device embedded in a moderator.

The calculations use the neutron Monte-Carlo code MONK<sup>(1)</sup> to predict the neutron flux in each region of a defined system. The merits of the common moderators  $H_2^0$ ,  $CH_2^-$  and  $D_2^0^$ have been investigated as well as the properties of other materials such as lead, graphite and nickel. The calculations consider liquid samples in a variety of geometries and determine the optimum position for BF<sub>3</sub> or <sup>3</sup>He counters. Calculations on the neutron yields from spontaneous fission and ( $\alpha$ ,n) reactions in aqueous solutions have also been carried out. Many calculations have been carried out<sup>(2)</sup> but it is only possible to present a selection of the results in this report.

(a) A specific geometry



Fig. 24. A definition of the regions used in the calculation of neutron flux arising from a central pipe surrounded by a cylinder and moderator

Consider the system illustrated in Fig. 24. It consists of a 2.5 cm diam. pipe of liquid passing through the centre of a cylinder of moderator which is divided into regions for the purpose of determining the axial and radial variations of flux. Neutrons from a fission spectrum of the form,  $f(E) = 0.4527 \exp(-E/0.965) \sinh(\sqrt{2.29E})$ originate from within the volume of the central pipe, which extends beyond the moderator, and are tracked until they are captured or leave the system. The flux distribution for  $CH_2$  moderator, with a small cylinder of lead in the regions just outside the central pipe, is given in Fig. 25. The radial variation of thermal flux for CH2,H20 and D20 moderators is given in Fig. 26 for the central regions 1 to 7. There is very little difference between H20 and CH2 which is consistent with the fact that carbon and oxygen both have small

capture cross sections. The  $D_2^{0}$ , on the other hand, is not such a good moderator and its moderating power is not able to compete below the radial distance of 15 cm. Beyond 15 cm it is superior to either CH<sub>2</sub> or H<sub>2</sub>0 because of the very low absorption cross section of deuterium. In the case of a 20 cm radius cylinder, 99.2% of the neutrons escape through a  $D_2^{0}$  moderator compared with 59% for CH<sub>2</sub> and 63% for H<sub>2</sub>0.



Fig. 25. Energy distribution of flux. Energy Fig. 26. Radial variation of thermal flux groups of equal lethargy from group 1 thermal to group 16 above 10.0 MeV

The response of a  $BF_3$  counter was determined for a 31EB70 tube\* at a radial distance corresponding to region 4. Neutrons were tracked through the system and the number captured in  ${}^{10}B$  was recorded. A similar technique was used to assess the performance of a  ${}^{3}$ He tube, type 31 He ${}^{3}$ /304/25B\* and a  ${}^{235}U$  fission counter, type FC165A/1000/235\*. The amount of  ${}^{3}$ He and  ${}^{235}U$  was assumed to be uniformly distributed throughout the volume of the counter and the argon/CO<sub>2</sub> gas filling was ignored. The neutrons were taken to originate in an 80 cm long, 3.0 cm diameter cylinder of water. The neutron counter

\*Twentieth Century Electronics Ltd.

### Table 1 Counts per neutron released from

the source

teren en e	
BF <sub>3</sub> (31 EB 70)	.007
<sup>3</sup> He (31 He3/304/25B)	.012
<sup>235</sup> U (FC165A/1000/235)	.00017

efficiencies for a single counter at a radius of 10.0 cm are given in table 1. They are expressed as counts per neutron released from the source. The calculations have also shown the extent to which the presence of a counter influences the flux. There is quite a severe depression of the flux for a <sup>3</sup>He counter which absorbs about 40% of the neutrons entering the counter

and gives rise to a "neutron shadow" on the side of the counter away from the source. A 5.0 cm diameter  $BF_3$  counter showed only a factor of two increase in efficiency over that of a 2.5 cm diameter counter. The efficiency should therefore be considered as being dependent on the solid angle for interception of neutrons.

(b) <u>A general consideration</u>



Fig. 27. Illustrating the effect of extra moderator behind a  $BF_3$  counter

The backscattering effect of  $CH_2$ moderator and its influence on a  $BF_3$  counter is illustrated in Fig. 27, where the effect of increasing the thickness of moderator behind the counter has been calculated. The neutrons originate from a cylindrical volume of radius 5 cm and length 31 cm. A moderator thickness of 10 cm is adequate for a  $BF_3$ counter placed just outside the source.

 V. S. W. Sherriffs, unpublished information

(2) B. W. Hooton, AERE R-8976

### A model for studying coincidence systems used in plutonium assay (B. W. Hooton and E. W. Lees)

The determination of plutonium by measurement of time correlated neutrons consists of measuring coincidences between neutrons emitted from spontaneous fission. It is a well established technique  $^{(1),(2)}$  but it is subject to certain limitations. The limitations are primarily due to four factors.

- 1. The presence of neutrons from  $(\alpha, n)$  reactions. This random component is usually present with unknown intensity since the chemical composition is generally unknown.
- 2. Neutron multiplication in the source. Spontaneous fission neutrons can induce further fission in the source material which gives rise to an effective  $\overline{\nu}$  much greater than that of spontaneous fission.
- 3. Problems associated with high counting rates.
- 4. The particular limitations of the system of electronics used to analyse the time correlated component of the neutron flux.

A model has been constructed to assess the merits of the various electronic systems proposed, and to study their capability for dealing with the limitations given above. The model produces a pulse train of neutrons each designated by a time coordinate. The pulses in this train are organised into chronological order and may then be fed into any system of simulated electronics. The model specifies a system in the following terms.

- 1. Random neutron rate in the sample.
- 2. Spontaneous fission rate in the sample.
- 3. A  $\overline{v}$  distribution for neutrons from spontaneous fission.
- 4. A fission multiplication rate in the sample.
- 5. A  $\overline{v}$  distribution for neutrons from fission multiplication.
- 6. The counting efficiency of the system.
- 7. The probability distribution for the time between emission of a neutron in the sample and detection. This distribution is parameterised by a linear rise time followed by an exponential die-away time.
- 8. A simulator for the electronic counting system is also specified.

The Monte Carlo technique is used to generate a time coordinate for each neutron detected by using a random number generator to make the following decisions.

1. The time coordinate of an event in the sample

- 2. The type of event, random, spontaneous fission or fission multiplication
- 3. The number of neutrons emitted
- 4. The number of neutrons detected
- 5. The time coordinate of each neutron detected

At high counting rates a problem can arise from overlapping events in the manner illustrated in Fig. 28. The long die-away time in the moderator surrounding the detectors can result in the detection time of a neutron from an early event being later than the detection time of a neutron from a later event. It is, therefore, necessary to queue the neutron pulses in correct chronological order before processing the pulses through an electronic system.











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The model has been used to study the properties of the Variable Dead-Time Counter (VDC) and particularly the Harwell version with  $BF_3$  counters in a moderator which has time characteristics of 5 µs rise time and 100 µs die-away. The results, with errors determined by considering the standard error associated with many groups of neutrons, show a departure from linearity (Fig. 29) for samples greater than 50 g of  $^{240}$ Pu. The results can also be erroneous if the percentage of ( $\alpha$ ,n) counts varies across the samples.

The difficulty in extracting true from accidental coincidences at rates of 6000 counts/s can readily be appreciated by the fact that a queue of five neutrons was barely adequate to maintain chronological order in pulses processed within the model. This observation shows that in a counting system with a die-away time as long as 100  $\mu$ s one can expect to get more than five overlapping events in the counting system at rates of 6000 counts/s.

The model will continue to be used to investigate properties of the VDC system and will be extended to examine the limitations of the shift register technique<sup>(3)</sup>.

- (1) K. P. Lambert and J. W. Leake, AERE R-8300 (1976)
- (2) R. Berg et al, EUR 5158e (1974)
- (3) K. P. Lambert, AERE R-8303 (1977)

## Calculations of neutron multiplication and its effect on the response of the Euratom VDC (E. W. Lees and B. W. Hooton)

Theoretical responses have been calculated for the Variable Deadtime Counter (V.D.C.) currently used by Euratom safeguards inspectorate to determine Pu mass by measurement of the <sup>240</sup>Pu spontaneous fission (SF) rate. The analysis of the VDC data has been assumed to follow that proposed by Stanners<sup>(1)</sup>.

The VDC essentially measures the neutron multiplicity from a fission event as detected by the counter assembly. This principle is identical to that employed in the analysis of neutron yield data from photofission and photoneutron experiments (PR/NP 24, p 39) and so the existing formalism and computer programs are simply modified to describe the Euratom VDC assembly. The neutron multiplication in samples of varying mass, density and geometrical arrangement was estimated using the Monte-Carlo program MONK<sup>(2)</sup> which lists the number of fast fission (FF) events induced by the neutrons from the original <sup>240</sup>Pu spontaneous fission neutrons. Since the FF occurs within a few  $\mu$ s of the original SF, then these events are essentially coincident in time and appear to the VDC as a

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single fission event with a much larger average number of neutrons being emitted than for  $SF(i.e. \bar{v}_{eff} = \bar{v}_{SF} + \bar{v}_{FF} - 1$  where  $\bar{v}$  is the average number of fission neutrons for each separate event). Since the probability of detecting 2 or more neutrons from a given event as a function of  $\bar{v}$  is highly non-linear, those events combining SF + FF will have a larger effect on the measured VDC response e.g. if 5.4% of the original SF events result in SF + FF then the response of the VDC is increased by 17.6%.



Fig. 30. Increase in VDC response due to neutron multiplication in spheres of increasing masses of Pu. In the absence of neutron multiplication, the VDC response is normalised to unity

Fig. 30 indicates the increase in response of the VDC as a function of mass for Pu0<sub>2</sub> containing approximately 20% of <sup>240</sup>Pu arranged in spherical geometry. As one would expect, isotopic and chemical composition, geometrical arrangements and density are important in calculating the increased response resulting from neutron multiplication. Fig. 31 indicates the effect of density for a fixed mass of 1 kg Pu0<sub>2</sub> arranged in spherical geometry, but of 'varying density. The density variation appears to be more important than geometrical considerations.



Fig. 31. Increase in VDC response due to neutron multiplication in an 1 kg sphere of Pu0  $_2$  as a function of density

The present calculations are being prepared for publication and future work will be directed to investigate neutron multiplication effects on the shift register technique of analysis<sup>(3)</sup>.

- (1) W. Stanners, Euratom Inspectorate, Luxembourg, private communication
- (2) V. S. W. Sherriffs, unpublished information
- (3) K. P. Lambert, AERE R-8303 (1977)

### A water tank study of neutron systems (B. W. Hooton, E. W. Lees and A. Talbot)

The water tank shown in Fig. 32 has been used for measurements of the thermal flux distribution of several sources and for experimental confirmation of calculations on passive counting systems for Pu assay (see this report p 60).



Fig. 32. Water tank used for measurements to verify design calculations

The radial distribution of thermal flux has been measured for sources of  $^{252}$ Cf, Am/Li, Am/B, and Pu/Be using BF<sub>3</sub> and <sup>3</sup>He counters. The distribution for  $^{252}$ Cf is given in Fig. 33 together with the results of a calculation using MONK<sup>(1)</sup>. Also presented in Fig. 33 is the theoretical distribution to be expected from a monoenergetic source of 10.0 MeV. The radial distributions for the other sources are given in Fig. 34.

The neutron die-away time characteristics of the system were investigated and are shown in Fig. 35. They were obtained using a start pulse from detection of a fission fragment from  $^{252}$ Cf to operate a time-to-amplitude converter and a stop pulse from detection of a neutron in a BF<sub>3</sub> counter. The large water bath produces a very long rise time and die-away characteristic. This distribution will be used to test calculations made using a time dependent version of MONK when the program modifications are complete.



Fig. 33. Radial distribution of thermal flux. Measurements with a BF<sub>3</sub> counter compared with Monte Carlo results from MONK









One important parameter in the design of neutron counting systems is the distance between adjacent  $BF_3$  or <sup>3</sup>He detectors. The influence of counters on their neighbours when the distance between them is reduced has been investigated and the maximum reduction in counting rate for two adjacent BF3 counters of 2.5 cm diameter was found to be 9%. The comparable figure for <sup>3</sup>He counters was 15%. It should be pointed out, however, that in a multicounter system the interference effects are due to many-body interactions. The water tank has been calibrated with standard sources and used to 1, 21 assay plutonium samples. The measurements have confirmed calculations on the  $(\alpha, n)$ 

yields from plutonium in aqueous solutions.

### (1) V. S. W. Sherriffs, unpublished information

### Assessment studies of nuclear incineration (F. Duggan\* and M. G. Sowerby)

The assessment studies on nuclear incineration previously reported (PR/NP 24, p 28) have been continued. The first phase of the calculations of single isotope incineration have been reported<sup>(1)</sup>. These studies have been concerned with assessing (i) the rate of conversion of the various actinides to fission products, (ii) the neutron balance of the system during incineration and (iii) the relative toxicity associated with the recycling of the actinides. In table 1 the neutrons produced per neutron absorbed during incineration are presented. A characteristic of the actinides is that the neutron yield is still significant even at high burn up. In Table 2 typical examples of the variation of Toxicity Index<sup>4</sup> with cooling time of the total actinide inventory produced by the burn up of the initial isotope is shown. It has now been pointed out by McKay<sup>(2)</sup> that the removal of the Pu/U produced during incineration is an inevitable component of the actinide incineration cycle. Therefore in the second phase of this work all the above calculations are being revised to include the recovery of Pu/U prior to assessing the toxicity.

In their review of the state of the art of nuclear incineration McKay et al<sup>(3)</sup> identified the investigation of the fuel cycle consequences of different nuclear incineration strategies as being a little studied but crucial area of work. In order to make further studies in this field the computer program HYLAS<sup>(4)</sup> is being extended so that actinide recycle calculations can be performed and the following factors considered:

- (a) the detailed effect of losses of actinides from the fuel cycle
- (b) the recovery of Pu/U during the reprocessing of fuel used for nuclear incineration

The toxicity index ( $\Phi$ ) for air or water is defined for a mixture of nuclides as

### $\Phi = \sum_{i} (Q_{i} / MPCP_{i})$

where  $Q_i$  is the activity in curies of the i<sup>th</sup> nuclide and MPCP<sub>i</sub> is the maximum permissible concentration of the nuclide in air or water as given by ICRP for the general public (i.e. one tenth of the occupational level for a 168 h week)

\*Imperial College, University of London

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Starting	Neutrons produced per neutron absorbed											
Nuclide		LWR		FBR			"DFR"*			HTR		
Irradiation time (y)	0	10	20	0	10	20	0	10	20	0	10	20
232 <sub>Th</sub>	0.00	0.88	0.89	0.04	1.31	1.30	0.20	1.41	1.42	0.003	1.02	1.03
<sup>231</sup> Pa	0.01	1.04	1.22	0.44	1.55-	1.04	1.21	1.93	2.05	0.01	1.02	1.24
<sup>234</sup> U	0.02	1.02	0.86	0.89	1.25	1.28	1.82	1.92	1.86	0.03	1.05	0.96
<sup>236</sup> U	0.03	0.51	0.64	0.39	125	1.35	1.32	1.66	1.71	0	0.45	0.57
<sup>238</sup> U	0.13	0.98	1.05	0.41	1.45	1.47	1.12	1.88	1.89	0.03	0.91	0.89
237 <sub>Np</sub>	0.01	0.81	0.82	0.44	1.93	1.72	1.42	2.30	2.22	0.01	0.77	0.77
238 Pu	0.12	0.91	0.87	2.05	1.88	- 1.64	2.63	2.53	2.16	0.13	0.88	0.72
<sup>239</sup> Pu	1.88	0.57	0.95	2.30	1.83	1.80	2.82	2.59	2.44	1.83	0.41	0.73
241 Am	0.02	0.89	0.93	0.80	1.85	1.71	1.56	2.29	2.32	0.04	0.85	0.75
<sup>243</sup> Am	0.05	1.21	1.15	0.64	1.72	1.56	1.24	2.49	2.61	0.01	0.87	1.01
<sup>244</sup> Cm	0.06	1.23	1.13	1.79	ľ.77	1.10	2.83	2.38	2.63	0.06	1.16	1.06
				•								

### Neutrons produced per neutron absorbed (n) during Nuclear Incineration

\*The same flux  $(10^{16} \text{ n/cm}^2/\text{s})$  was used for DFR and FBR and this implies a lower power density for DFR by  $\sim 30\%$ 

Table 1

Starting	Time after	LI	WR.	]	FBR	"I	)FR''*	H	rr
Nuclide	(y)	f <sub>W</sub>	fA	f <sub>W</sub>	fA	f <sub>W</sub>	fA	f <sub>W</sub>	fA
237 Np	10 <sup>3</sup>	0.06	0.04	1.5	0.98	7.2	4.4	0.04	0.03
1	10 <sup>4</sup>	0.10	0.1	5.4	3.5	6.8	12.8	0.06	0.09
	10 <sup>5</sup>	0.13	3.1	9.9	322	5.9	255	0.03	1.9
	10 <sup>6</sup>	1.4	1.3	117	1687	71	1800	1.0	7.4
	107	11.7	16.2	1252	2219	1365	3137	6.9	8.9
	10 <sup>8</sup>	$4 \times 10^{-11}$	$2 \times 10^{-10}$	3x10 <sup>-9</sup>	2x10 <sup>-8</sup>	8x10 <sup>-9</sup>	$1.5 \times 10^{-8}$	$3 \times 10^{-11}$	1x10 <sup>-10</sup>
243 <sub>Am</sub>	10 <sup>3</sup>	3 0	13	24	14	73	33	17	0.7
7111	10 <sup>4</sup>	4.9	3.1	40	33	116	74	2.6	1.6
	10 <sup>5</sup>	3.9	15	32	81	86	414	2.4	7.5
	10 <sup>6</sup>	0.06	0.07	0.37	0.37	0.99	1.6	0.04	0.07
	107	0 <b>.</b> 87	2.9	5.6	12.2	16.8	30	0.59	1.7
	10 <sup>8</sup>	1.93	46.7	14.5	222	100	510	1.6	9.7

### Reduction factors for the toxicity indices: $f_W$ (water) and $f_A$ (air) produced by a 20 year irradiation in various reactor spectra

Table 2

 $f_W$  and  $f_A$  are dimensionless quantities which give the toxicity reduction with respect to the toxicity of the initial isotope. When  $f_W, f_A^{< 1}$  an increase in toxicity is implied

\* The same flux  $(10^{16} \text{ n/cm}^2/\text{s})$  was used for DFR and FBR and this implies a lower power density for DFR by ~30%

-85 - (c) the net consumption of Pu/U during nuclear incineration

(d) the need and toxicity of a possible store for higher actinides.

The program is being written so that it is possible to add and remove actinide isotopes at any time during the recycling. The modified program is not yet fully operational but test cases are being run.

All the material presented at the First Technical Meeting on the Transmutation of Actinides, Ispra, March 1977 is being collated in order to keep the literature survey up to date.

- F. Duggan and A. J. H. Goddard, Proc. of First Technical Meeting on the Nuclear Transmutation of Actinides, Ispra (1977) EUR 5897 p 263
- (2) H. A. C. McKay, Proc. of First Technical Meeting on the Nuclear Transmutation of Actinides, Ispra (1977) EUR 5897 p 3
- H. A. C. McKay, M. G. Sowerby, M. Bustraan, J. Montizan, A. Van Dalen and B. Verkerk, EUR 5801e (1977)
- (4) R. H. Clarke, CEGB Report RD/B/N2118

### Fuel cycle studies (M. G. Sowerby and T. W. Conlon)

The present U.K. fuel cycles for thermal and fast power reactors have evolved over a period of more than 20 years. Many of the major decisions regarding these cycles were made at the start of this period when environmental aspects did not carry their present weight and our knowledge was not as good as it is today. It was felt therefore that it was now opportune to start a fuel cycle optimisation exercise whose objective was to optimise the nuclear fuel cycle with respect to all known constraints but with particular emphasis on environmental aspects. The ideas behind this programme originated at Harwell and the Chemistry, Chemical Technology, Environmental and Medical Sciences, Metallurgy and Nuclear Physics Divisions are all contributing to the work. The contribution from Nuclear Physics Division came from the present authors. From the beginning it was recognised that much help from other U.K.A.E.A. establishments would be required and this is now being given by staff based at London Office, Risley, SRD (Culcheth) and Winfrith.

The work being undertaken can be divided into 3 main parts:-

 The reasons for choosing the present U.K. strategy on the fuel cycles for thermal and fast reactors are being compiled and factors which limit possible modifications to the fuel cycles are being identified;

(2) A method is being developed for quantifying the "hazard" (i.e. the environmental

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- impact in terms of radiation dose to man) of the fission reactor fuel cycle;
- (3) Changes to the fuel cycle which might reduce the "hazard" are being proposed and investigated.

## Development of the fission product inventory code FISPIN (B. W. Thomas and D. A. J. Endacott)

The fission product inventory program FISPIN, developed originally by Richardson<sup>(1)</sup>, calculates inventories of fission products, actinides and activation products during and after irradiation in a nuclear reactor. Additional information is also provided concerning the heat output and the nature of the radioactive activity of the isotopes.

<sup>'</sup> Development of FISPIN in the past has involved many program alterations which led inevitably to a generally untidy and cumbersome code which existed in several different forms within the U.K.A.E.A. Major improvements were made recently by R. B. Thomas<sup>(2)</sup> with a view to establishing an acceptable version for general use.

Nuclear Physics Division has now taken on responsibility for the Harwell version of the code (available on the IBM 370 computer) and will be directly involved in the future development of new facilities. Substantial progress has been made in eliminating certain errors in the original program and some effort has been devoted to the establishment of a series of direct access data libraries on disc.

Work in the immediate future will be directed towards removing the few remaining faults in the code and eliminating anomalies between the different versions within the U.K.A.E.A. Planned extensions to the code include (i) ability to read library data in the ENDF/BIV form, (ii) hazard calculations and (iii) graphical output.

- (1) B. L. Richardson, unpublished information
- (2) R. B. Thomas, AERE M 2872 (1977)

### Data files for the U.K. nuclear data library (Miss E. M. Bowey and Mrs. C. M. Chaffey)

Work on the progress referred to in PR/NP 24, p 30 has been completed and a satisfactory set of programs is now available on the Harwell computer to allow compilation of new data files and amendment of existing files when required.

In addition the program MINIGAL<sup>(1)</sup>, used to calculate average cross-sections and resonance integrals from files in the U.K. Nuclear Data Library is now available at Harwell.

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The new data file for  $^{238}$ U is complete. This includes a recent evaluation of the fission cross-sections. A file for  $^{241}$ Am has been constructed and is virtually complete, using the data of Lynn (PR/NP 23, p 27). Work on the  $^{243}$ Am file is in progress.

(1) A. L. Pope and J. S. Story, Winfrith report AEEW - M 1191 (1973)

#### Assessment of transmutation and activity in fusion reactors (0. N. Jarvis)

The computer code ORIGEN has been selected as the most useful vehicle for the study of the activation of materials likely to be found in the structure of a fusion reactor. ORIGEN<sup>(1)</sup> is a card-input orientated program intended for fission reactor studies: it has been modified for convenient use at the local IBM 370/165 computer and extended to cater for the wider range of nuclear reactions of interest with a 14 MeV neutron source. The current situation relating to the nuclear data libraries is as follows:-

- (i) A new  $\gamma$ -ray library has been constructed from the Jülich compilation of Erdtmann and Soyka<sup>(2)</sup>.
- (ii) A comprehensive cross-section library has been specified, containing all nuclides likely to be produced from structural materials and impurities. This library is prepared in a spectrum-averaged form for direct access by ORIGEN.
- (iii) The spectrum averaged library is constructed at present from a 100-group cross section library tape DLC-33<sup>(3)</sup> which contains only a fraction of the required reactions.
- (iv) Data for the important stable nuclides are accessible from ENDF B/IV.
- (v) THRESH2<sup>(4)</sup> has been used to generate synthetic multigroup data for unstable nuclides and for those stable nuclides not listed in ENDF B/IV.
- (vi) Neutron capture  $(n,\gamma)$  reactions are not provided by THRESH2. To rectify this deficiency the program FISPRO<sup>(5)</sup> is being investigated.

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(1) M. J. Bell, ORNL-4628 (1973)

- (2) G. E. Erdtmann and W. Soyka, Nucl. Inst. & Meth. 121 (1974) 197
- (3) Available from the RSIC Data Library Collection, ORNL
- (4) S. Pearlstein, J. Nucl. Energy 27 (1973) 81
- (5) V. Benzi and G. Reffo, CCDN-NW/10 (1969)

#### CHEMICAL NUCLEAR DATA

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### Introduction

Chemical nuclear data experiments and evaluations are coordinated by the Chemical Nuclear Data Committee (Chairman: J.G. Cuninghame). This committee, which is also the Chemical Sub-Committee of the UKNDC, is made up of measurers, evaluators and users of chemical nuclear data and is able to advise on measurements and to consider in detail reports of work on the compilation and evaluation of data in this field. The preparation of the CNDC nuclear data file is under the control of the Data File Sub-Committee (DFSC) whose new chairman is Mr. B.S.J. Davies (CEGB, BNL). The main committee held two meetings and the sub-committee three meetings in 1977. The request list was revised early in the year (CNDC (77) Pl) and is due to be published in conjunction with the 'Physics' list. Work is in progress on very nearly all items on the list but often on a long time scale and with little prospect of early completion. Data File work, however, has now reached the stage where data is available to the user and the contents of the file are listed below.

The efforts of the committee to reduce the duplication of work have been successful and the objective of effective international cooperation in the data field is being pursued.

This section has been compiled and edited by I.C. McKean (Secretary - CNDC, AERE) and brings together reports on the chemical nuclear data work being done at Harwell (AERE), Winfrith (AEEW), Aldermaston (AWRE), Dounreay (DNPDE), Windscale (BNFL) and Berkeley (CEGB) under the headings:

#### (A) MEASUREMENTS

- 1. Fission Yields
- 2. Neutron Cross Sections
- 3. Half-lives
- (B) EVALUATIONS AND COMPILATIONS
  - 1. Data File Sub-Committee
  - 2. Fission Yields
  - 3. Related Compilation Studies
- (A) MEASUREMENTS
- 1. Fission Yields

### Mass spectrometric measurements of alpha in PFR (I.C. McKean, E.A.C. Crouch, J.G. Cuninghame, H.H. Willis (AERE), with V.M. Sinclair, W. Davies (DNPDE) and N.R. Daly (AWRE))

The samples which were prepared in 1973 are now being irradiated in PFR.

### Tritium yields in thermal and fast fission (I.C. McKean, E.A.C. Crouch (AERE)

DIDO rabbit irradiations of  $^{235}$ U in aqueous solution contained in polythene containers have given  $^{3}$ H/fission ratios of 0.93 x 10<sup>-4</sup>, 0.87 x 10<sup>-4</sup> and 0.95 x 10<sup>-4</sup>. These (provisional) figures average to 0.92 x 10<sup>-4</sup> tritons perfission (t/<sub>f</sub>) compared with the mean from existing chemical method results in the literature of (0.89 ± .04) x 10<sup>-4</sup> t/<sub>f</sub>.

Because <sup>239</sup>Pu cannot be irradiated in a similar manner in the 'DIDO' rabbit due to safety considerations, preparations are being made to do the irradiations in 'GLEEP', together with repeat irradiations of <sup>235</sup>U.

Water and polythene cannot be used in 'ZEBRA' irradiations because of their effect on the neutron energy spectrum and so an attempt will be made to use super-pure Al containers for the fast neutron irradiations. It is thought that this material will contain a small enough amount of Li to allow its effects to be measured by spiking.

#### Fission yield measurements at DNPDE (W. Davies, V.M. Sinclair (DNPDE))

Of the reactor irradiations described on pages 43 and 44 of the 1974-75 UNKNDC Progress Report, changes in emphasis in the work programme made it unnecessary to proceed with the DFR experiment and work on it ceased in March 1977.

The PFR experiment is still in hand, but no further progress has been made during the past year.

# Absolute fast fission yields of <sup>99</sup>Mo and other nuclides in <sup>238</sup>U fission (J.G. Cuninghame, H.H. Willis (AERE))

This work is half completed but is at a standstill because of staff shortage. <u>Absolute fast fission yields of <sup>99</sup> Mo and other nuclides in <sup>239</sup> Pu fission (J.G. Cuninghame,</u> H.H. Willis (AERE))

Work complete and published; J. Inorg. Nucl. Chem. 39 (1977) 383.

## ZEBRA-BIZET experiments to study the effect of change of reactor neutron spectrum on fission yields (J.G. Cuninghame, H.H. Willis (AERE))

Partly as a result of the continued failure to obtain low power irradiations in PFR and partly because of the success of experiments in the  $\gamma$ -analysis of unseparated fission products, it has been decided that measurements of the effect of change of neutron spectrum on fission yields will be made mainly by experiments in the ZEBRA-BIZET core. We hope to obtain fission yields for <sup>235</sup>U, <sup>238</sup>U and <sup>239</sup>Pu fission in four different fast reactor spectra in a series of monthly irradiations. The first such irradiation has been done and the samples are now being counted.

### Development of a method for the simultaneous measurement of fission yields of a large number of fission products from a low flux irradiation of nuclear fuel (J.G. Cuninghame, H.H. Willis (AERE))

One of the principal problems facing those who wish to make measurements of fission yields and other fission product and actinide nuclear data for fast reactors, is the extreme difficulty of having samples irradiated to high total doses in the appropriate neutron spectrum. This means that the normal mass-spectrometric or high intensity  $\gamma$ -counting analytical procedures are frequently unavailable and the only irradiation facilities are accelerators or zero energy reactors for which the practical total neutron dose for a sample is in the range  $10^9 - 10^{12}$ . Given such low neutron fluxes the usual analytical technique employed has been  $\beta$ -counting of thick sources prepared after laborious and time-consuming radio-chemical separation and purification.

We are developing techniques for  $\gamma$ -counting of fissile material with little or no chemical separation, so that all fission products with suitable  $\gamma$ -rays are measured simultaneously. The  $\gamma$ -spectrometers have been accurately calibrated and the ultimate aim is to be able to obtain absolute measurements of the yields of at least 15 fission products with  $\gamma$ -counting rates in the photo peaks sometimes as low as 1 count/min., with an absolute accuracy for a fission yield of from 3-10%, depending on count rate.

In test irradiations in which only about  $10^{11}$  fissions were generated in the target material we have successfully identified up to 120  $\gamma$ -ray peaks belonging to 30 fission products from the total  $\gamma$ -ray spectrum of the untreated target and obtained satisfactory decay curves from the majority of them.

## Development of a version of the GAMANAL computer programme primarily applied to fission yield studies (J.G. Cuninghame, J.A.B. Goodall (AERE))

GAMANAL is a sophisticated computer programme running on the IBM 370 for fitting all the photopeaks in a 4096 channel spectrum. The programme first finds the peaks, fits them, applies appropriate calibration data, and finally lists all peaks found, together with their energies, count rates, and error data.

The version we received from Kaufman of the Argonne National Laboratory performed only these functions and required input from cards or IBM magnetic tape. This programme has been developed to provide the following additional facilities (some of which are optional):

- (i) input from Dectape from both our PDP8 and PDP11 spectrometers;
- (ii) input from floppy discs from our PDP11 spectrometer;
- (iii) plotting of all peaks on the line printer with original data, fit and continuum;

- (iv) full spectrum plot on the line printer with original data, fit and continuum;
- (v) similar full spectrum plot on the Calcomp or Versatek in 4 frames;
- (vi) Calcomp or Versatek plot of selected peaks in 1 frame to show all the separate Gaussian elements of the fit;

(vii) output on cards suitable for use as input to a decay curve analysis programme. Development of a computer programme to extract half-life data from  $\gamma$ -spectra analysed by GAMANAL (J.A.B. Goodall (AERE))

A card option has been added to GAMANAL which outputs all analysed  $\gamma$ -ray peaks, one per card, and includes all other data required for analysis of decay curves of up to 6 half-lives.

The sets of cards for the peaks to be analysed are input to this new programme which then performs a multiple exponential stripping calculation on the data, from which it calculates the zero time activities of all the components.

### Development of software to allow a PDP11 computer to perform as four independent, automatic 4-K pulse height analysers (J.G. Cuninghame, J.A.B. Goodall, A.L. Nichols, H.H. Willis (AERE) and members of Nuclear Enterprises Ltd))

A 28-K PDP11 computer, equipped with floppy disc and Dec-tape, is interfaced via Camac to up to four detection systems. 16-K of core is used as data for four 4-K pulse height analysers.

The software allows all four analysers to operate completely independently under either manual or automatic control. Quite extensive off-line data processing can be carried out on archived data using one of the analyser data areas while the other three continue with normal data acquisition; more sophisticated data processing is done by GAMANAL with data input direct from the PDP-11 Dec-tapes.

Future developments include storage and processing of data on floppy discs and remote control of any of the four analysers, e.g. by pulses from the Variable Energy Cyclotron (VEC).

An investigation of angular momentum distribution of primary fission fragments in <sup>208</sup>Po\* and <sup>209</sup>Bi\* fission by radio-chemical measurement of isomer ratios (J.A.B. Goodall (AERE) C. Branquinho, J. Freeman, J. Hemingway, G.W.A. Newton and V.J. Robinson of Manchester University)

Information about angular distribution of primary fission fragments can be obtained by measuring the ratio of metastable to ground states for pairs of isomers formed in fission. The pairs studied in this work were  ${}^{82}$ Br,  ${}^{83}$ Br,  ${}^{120}$ Sb,  ${}^{126}$ Sb and  ${}^{128}$ Sb, produced in the fission experiment described in the next item, in separate VEC ( ${}^{4}$ He +  ${}^{204}$ Pb) irradiations at Harwell, and in ( ${}^{12}$ C +  ${}^{209}$ Bi) irradiations on the Manchester Hilac. Isomer ratios have been predicted for fragments whose angular momentum distribution is taken from statistical theory assuming that the fissioning nucleus reaches the scission point along a low viscosity path.

Table 1 gives examples of the predicted values compared with the experimental ones. This work is now being prepared for publication and preparations have been made for extending it to the fission of highly fissile compound nuclei.

verage	Standard			
ragment	Deviation,	Calculated	Fragment	Experimental
Spin, h	h	Isomer Ratio	Studied	Isomer Ratio
20	5.97	1.54 <u>+</u> 0.41	<sup>120</sup> Sb	2.13 <u>+</u> 0.51
23	6.86	4.80 + 0.95	126 <sub>Sb</sub>	5.02 + 0.49

### Table 1: Predictions of the Fragment's Spin Distribution

FOI CHE	UI (0) Hev	C, IISSION) Rea	CLION	
19	5.67	1.38 <u>+</u> 0.35	<sup>120</sup> sb <sup>2</sup>	1.62 <u>+</u> 0.49
21	6.26	4.02 <u>+</u> 0.7	<sup>126</sup> Sb	4.40 <u>+</u> 0.18

### Mass distribution and charge dispersion in <sup>208</sup>Po\* fission measured by radiochemical means (J.A.B. Goodall (AERE) J. Freeman, J. Hemingway, G.W.A. Newton and V.J. Robinson of Manchester University)

This work is the radiochemical analogue of the next item of this report, the same targets and beams being used for measurements. Work on the systems ( ${}^{4}$ He +  ${}^{204}$ Pb) and ( ${}^{12}$ C +  ${}^{196}$ Pt) is now complete while the study of ( ${}^{16}$ O +  ${}^{192}$ Os), earlier held up because of target preparation problems, is in progress.

Relative mass yields have been measured for different isotopes of a number of elements and the most probable mass  $A_p$  and the isobaric width  $\Gamma_z$  are derived from these. The isobaric width must be the same for complementary elements at the moment of fission (complementary elements are pairs for which the sum of the atomic numbers is equal to the atomic number of the fissioning nucleus). The values of  $\Gamma_z$  estimated so far show a marked increase with Z, an increase which must be due either to neutron evaporation from the primary fragments or to multi-chance fission. Evaporation calculations show that neutron

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emission from the primary fragments does not, in fact, contribute much to  $\Gamma_z$  but almost all the observed width can be accounted for by multi-chance fission. This implies that the charge dispersion of primary fragments must be extremely narrow, with  $\Gamma_z$  comparable to the values found in spontaneous and thermal neutron induced fission.

### On-line measurement of variation with energy and angular momentum of total kinetic energy and pre-neutron emission fission fragment masses in fission of <sup>208</sup>Po\* (J.G. Cuninghame (AERE), J. Durell, G. Foote and I.S. Grant of Manchester University)

This work is now complete and is being written up for publication.

On-lin	e measu	irement of	variation	s with	energy	and a	ngular	momentum of	total kinetic energy
and pr	e-neut:	ron emissi	on fission	fragme	ent mas	ses in	fissio	on of highly	fissile compound
nuclei	(J.G.	Cuningham	e (AERE),	J. Dure	ell, G.	Foote	and I	S. Grant of	Manchester University

The object of this work is to compare the behaviour of highly fissile nuclei with the  $^{208}$ Po\* used in the experiment above. We are studying the reactions:

$$232_{\text{Th}} + {}^{1}_{\text{H}} \rightarrow 233_{\text{Pa}}$$

$$209_{\text{Bi}} + {}^{15}_{\text{N}} \rightarrow 224_{\text{Th}}$$

$$206_{\text{Pb}} + {}^{18}_{0} \checkmark 224_{\text{Th}}$$

The experimental work on the first two reactions is now complete.

Measurement of elastic scattering and fission cross sections for fission of <sup>208</sup>Po\* (J.G. Cuninghame (AERE), J. Durrell, G. Foote and I.S. Grant of Manchester University)

We are measuring elastic scattering and fission cross-sections in the reactions  $({}^{12}C + {}^{196}Pt)$  and  $({}^{16}O + {}^{192}Os)$  and we hope also to do so for the reaction  $({}^{4}He + {}^{204}Pb)$ . So far we have completed the measurements at three different excitation energies for the first reaction and at two for the second. We also intend to extend the measurements to the highly fissile systems referred to above.

The measurement of  $\overline{\nu}$  as a function of fission fragment mass and energy in fission of highly fissile compound nuclei (J.G. Cuninghame (AERE), J. Durell, G. Foote and I.S. Grant of Manchester University)

The measurements are over half completed.

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The measurement of  $\overline{\nu}$  as a function of fission fragment mass and energy in fission of  $\frac{208}{Po*}$  (J.G. Cuninghame (AERE), J. Durell, G. Foote and I.S. Grant of Manchester University)

Although these physical measurements are complete they are partially interlocked with the radiochemical ones and the calculations cannot be completed until the  $(^{16}0 + ^{192}Os)$  radiochemical work is finished and certain normalising parameters have been derived from

## The competition between fission and neutron emission in Po compound nuclei (J.G. Cuninghame (AERE), J. Durell, G. Foote and I.S. Grant of Manchester University)

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We have studied the competition between fission and neutron emission in the reaction  $^{204}\text{Pb}$  at bombarding energies from 50 to 80 MeV, by observing prompt  $\gamma\text{-rays}$  from the .α<sup>`</sup>+ Po isotopes. We measured excitation functions for the  $6 \rightarrow 4 \rightarrow 2 \rightarrow 0$  cascade of the eveneven isotopes <sup>204</sup> Po and <sup>202</sup> Po. In the case of <sup>202</sup> Po the ground state band had previously been observed only in heavier ion reactions at a single energy: the previous assignments are confirmed by our observations. Excitation functions for the  $2 \rightarrow 0$  transitions are shown in Fig. 1. together with the predictions of the evaporation code ALICE, using a level density parameter A/10. The same parameter gives a good account of the total fission cross section in ( $\alpha$  + <sup>208</sup>Pb), shown in Fig. 2. The good agreement gives confidence in the code's estimate of the probabilities of the average number of pre-fission neutrons, which increases from 0.6 to 2.4 as the bombarding energy increases from 50 to 80 MeV.



Fig. 1 Excitation functions for 2  $\rightarrow 0$  transitions in  $^{202}\text{Po}~(\clubsuit)$  and  $^{204}\text{Po}~(\clubsuit)$  with the predictions (solid lines) of the evaporation code ALICE using a level density parameter A/10.

We hope to extend this work to the other Po\* systems studied and also to the highly fissile systems mentioned above.

Fig. 2 Total fission cross-section for the reaction 4He + 208Pb (and 4He + 206Pb from other work) with the predictions of the evaporation code ALICE.

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#### 2. Neutron cross sections

## Measurement of neutron cross sections in PFR (W. Davies, D.J. Lord and V.M. Sinclair (DNPDE))

Some progress has been made in the past year on the work outlined on pages 53 - 55 of the 1974-75 UKNDC Progress Report: irradiation of the sub-assemblies containing all the reaction rate experiments except the low-power experiments was commenced in PFR in March 1977, or earlier.

### DFR Irradiation of <sup>242</sup>Pu (V.A. Proudler, W. Davies, and V.M. Sinclair (DNPDE))

The capsule containing the irradiated <sup>242</sup>Pu has been dissolved and analysis of the solution for americium and curium isotopes has commenced.

### Measurement of the integral capture cross section of <sup>243</sup>Am (Mrs. K.M. Glover, Miss S.A. Hall (Vacation Student Assistant), I.C. McKean, R.A.P. Wiltshire (AERE))

Progress in this area was again severely restricted during the early part of the year due to lack of effort.

All the <sup>241</sup>Am and <sup>243</sup>Am samples irradiated in ZEBRA core 14, a close simulation of the PFR neutron spectrum, have now been reprocessed. The cross section for the production of <sup>242</sup>Cm from <sup>241</sup>Am has yielded a result of 1.275  $\pm$  0.013 barns which is in good agreement with the expected values. Provisional results for the production of <sup>244</sup>Cm from <sup>243</sup>Am suggest a value of 1.51 barns.

The <sup>243</sup>Am has been reprocessed and three samples have been irradiated in the ZEBRA-BIZET CFR type core. These are expected to yield a further set of values for the production cross section of <sup>244</sup>Cm.

# Integral capture cross section of <sup>241</sup>Am and <sup>243</sup>Am in PFR (Mrs. K.M. Glover, R.A.P. Wiltshire (AERE))

Irradiation of these samples is expected to be completed during 1978.

3. Half-lives

237<sub>Np</sub> half-life (Mrs. K.M. Glover, D. Brown (AERE))

No progress.

### 239 Pu half-life (D. Brown, Mrs. K.M. Glover, M. King, R.A.P. Wiltshire (AERE))

Final evaluation of the results has been held up due to lack of effort but completion is expected during 1978.

<sup>241</sup>Pu half-life-1 (E.A.C. Crouch, I.C. McKean, (AERE))

(a) Lounsbury/Hall/Crouch measurements on old <sup>241</sup>Pu

Measurements on a sample of <sup>241</sup>Pu at least 25 years old have been made by massspectrometer and the results below are expressed as the "ratio of ratios" ( $\frac{241-241}{240-242}$ ) so that any errors due to mass-spectrometer bias should cancel.

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Analyst	Date of Analysis	Time from original analysis	Ratio of Ratios 241 <sup>2</sup> /240.242
.(1) Lounsbury	7.11.52	0.yr.	•5697
(2) Hall	26.11.53	1.05 yr.	.5102
(3) Crouch	24. 2.70	17.30 yr.	.1059
(4) Crouch	14. 3.77	24.35 yr.	.05358

Table 2: Analytical Results on old 241 Pu

Using the relation

 $\ln(\frac{241}{240},\frac{241}{242}) = \ln(\frac{241}{240},\frac{241}{242})_0 + (\lambda_0 + \lambda_2 - 2\lambda_1)T$ 

the above analyses yield the following half-life results:

Lounsbury/Crouch	(1)-(3)	gives	$t_{1/2} = 14.25 \text{ yrs.}$
Lounsbury/Crouch	(1)-(4)	gives	$t_{1/2} = 14.26 \text{ yrs}.$
Hall/Crouch	(2)-(3)	gives	$t_{1/2} = 14.31 \text{ yrs.}$
Hall/Crouch	(2)-(4)	gives	$t_{1/2} = 14.31 \text{ yrs.}$
Crouch/Crouch	(3)-(4)	gives	$t_{1/2} = 14.32 \text{ yrs.}$

The error associated with each of these figures is approximately 0.10 yr. (10). (b) Crouch  $\frac{240}{241}$  ratio and  $\frac{241^2}{240}$ . 242 ratio of ratios measurements on new  $\frac{241}{Pu}$ 

Two samples of <sup>241</sup>Pu (Series A and Series B) were prepared by the irradiation of <sup>240</sup>Pu in a thermal reactor in 1970 and have been analysed by mass spectrometer at frequent intervals since that date. Series A

at February	1976	241 240	rat	io			gave	t½	н	14.24	yr.	<u>+</u>	.12
(Sample now	exhaust	ed)	$\frac{241^2}{240 \cdot 242}$	ratio	of	ratios	gave	tł	8	14.53	yr.	<u>+</u>	•12

#### Series B

at February 1977  $\frac{241}{240}$  ratio  $gave t_{2}^{1} = 14.53 \text{ yr.} \pm .08$  $\frac{241^{2}}{240.242}$  ratio of ratios  $gave t_{2}^{1} = 14.33 \text{ yr.} \pm .11$ 

### <sup>241</sup> Pu half-life - 2 (Mrs. K.M. Glover, A.J. Fudge, E.A.C. Crouch, I.C. McKean, C. Whitehead, M. Wilkins, (AERE), P. De Bievre, B. Rose, R. Vaninbroux (Geel))

The enigma of the half-life of <sup>241</sup>Pu remains unresolved. Independent measurements by De Bievre (Geel) on both the Crouch and Wilkins samples are in good agreement with the half-life values previously reported for each of these samples.

A meeting was held at Harwell on November 23rd 1977 to discuss further strategy with a view to looking into the possibility of <sup>241</sup>Pu having two isomeric states with similar half-lives. On the assumption that sufficient material remained, further measurements on both the Crouch and Wilkins samples by alpha and gamma growth were suggested.

(B) EVALUATIONS AND COMPILATIONS

### 1. Data File (decay schemes) Sub-Committee (B.S.J. Davies (CEGB), M.F. James (AEEW), A.L. Nichols (AEEW), J.R. Parkinson (BNFL), A. Tobias (CEGB) and D.G. Vallis (AWRE))

Compilation and evaluation of nuclear decay data has continued with an effort of approximately 1.5 manpower. The priorities are defined by the sub-committee and linked to the requirements of the UK reactor programme. In order to achieve consistency and for ease of comparison and use, the internationally accepted ENDF/B4 format has been adopted.

The UK CNDC data files now contain:

(1) The fission product decay set UKFPDD/1, which was constructed by merging the data due to Tobias<sup>(1)</sup> with the US ENDF/B4 file. This contains data for 837 fission product nuclides, including 120 stable nuclides and 301 for which complete spectral data are given. The data include values of half-life, decay energies, intensities and internal conversion coefficients, branching ratios, mean energies and Q-values, all with associated errors whenever possible.

- (2) A set of fission product yield values due to Crouch<sup>(2)</sup>. This contains values of yields for fission induced by neutrons with energies ranging from thermal to
   14 MeV and for 7 fissile nuclides.
- (3) A set of decay data for 91 activation products of structural materials due to Nichols. The parameters given are similar to those for fission products listed above.

A report describing the data for activation products is being prepared. Nichols has started evaluating decay data for a list of actinides requested by CEGB. The production of this file is continuing.

Work on up-dating the fission product decay data is in hand with the aim of producing a revised set by mid-1978.

Table 3 shows the status of the data file at the end of 1977.

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(1) A. Tobias, CEGB Report RD/B/M2669 (1973)

(2) E.A.C. Crouch, Atomic Data and Nuclear Data Tables, 19, (1977) 417

#### 2. Fission Yields

### Fission yield assessments and consistent sets (E.A.C. Crouch (AERE))

The assessments and the consistent sets mentioned in the last Annual Report have now been published in "Atomic Data and Nuclear Data Tables" <u>19</u>, (5), 1977, p.417. It will be recalled that the consistent sets were produced by adjustments which caused the sets to obey the constraints

Where Y(A) is the adjusted chain yield for fission products of mass A, FIY(A,Z) is the adjusted independent yield of nuclide mass A, atomic number Z and  $\overline{\nu}_r$  is the number of

### Table 3: UK Chemical Nuclear Data File in ENDF/B4 Format

### November 1977

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Item	Data	Present Status	File Development
1	α decay data	Completed in non-ENDF format by Rogers. Published as AERE-R8005.	No effort available for conversion to ENDF.
2	Fission Product decay data, in- cluding nuclides produced by acti- vation of F P's.	This is the main effort of the subcommittee. Major contributions are by BNFL (Parkinson) and CEGB (Davies, Tobias). Tobias' data from RD/B/M2669 have been merged with the US ENDF/B4 file to produce UK FPDD/1. At BNFL new evaluations have been done for Te 131, I 131, Xe 131m, Pr 144, Pr 144m, Cs 137, Ba 137m, and are in hand for Te 131m, Te 132, I 132, Cs 134, Cs 136, Ce 143, Pr 143, Ce 144. At CEGB new data sets have been derived from the ENSDF for about 80 nuclides.	The aim is to update UKFPDD/1 by June 1978.
3	Activation pro- ducts of struct- ural materials.	Completed by Nichols for list of nuclides requested.	Data in ENDF/B4 format are available for H3, Be 10, C 14, C 15, N 16, O 19, Na 22, Na 24, Na 24m, Mg 27, S 35, C1 36, C1 38, C1 38m, Ar 39, Ar 41, K 40, K 42, Ca 41, Ca 45, Sc 46, Sc 46m, Sc 47, Sc 48, Cr 51, Mn 54, Mn 56, Fe 55, Fe 59, Co 57, Co 58, Co 58m, Co 60, Co 60m, Ni 59, Ni 63, Ni 65, Cu 64, Cu 66, Zn 65, Br 82, Br 82m, Y 88, Nb 93m,Nb 94, Nb 94m, Mo 93, Mo 93m, Ag 110, Ag 110m, In 115, In 115m, In 116, In 116m, In 116m2, Sn 121, Sn 121m Sb 122, Sb 122m, Sb 124, Sb 124m1, Sb 124m2, I 126, I 128, Cs 134, Cs 134m, Cs 136, Cs 136m, Ba 133, Ba 133m, Pm 145, Eu 152, Eu 152m1, Eu 152m2, Eu 154, Eu 154m, Tb 157, Hf 175, Hf 181, Ta 182, Ta 182m1, Ta 182m2, W 181, W 185, W185m, W 187, Au 198, Au198m Hg 197, Hg 197m, Hg 203.

Table 3: Continued

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Item	Date ···.	Present Status	File Development
4	Heavy Element and actinide decay data.	Further evaluations are under way。	Data in ENDF/B4 format are available for Hg 206, Tl 206, Po 216, Rn 220, Ra 225, Ra 226, Ra 228, Ac 228, Th 228.
5	Decay data for other nuclides.	No effort available.	
6	Fission yields.	Crouch's second round of evaluations com- pleted and published in Atomic and Nuclear Data Tables 19, 5, p 417 (1977).	Data are available in ENDF/B4 format.
7	Delayed neutrons.	Tomlinson data are still recommended for use. New evaluation by Crouch underway。	
	Spontaneous fission data.	Will be covered by M. King and Mrs. Glover who will also maintain Rogers α-library.	•
9	X-ray energies and intensities, β+/EC conversion coefficients.	Values are being incor- porated into data for particular nuclides under items 2, 3, 4 as appropriate.	
10	Cross-sections of fission products.	No effort available.	

neutrons emitted per fission.  $Z_F$  is the atomic number of the fissile nuclide and  $A_F$  is its atomic mass. The adjustments were found using four simultaneous equations with four undetermined multipliers. Condition (4) above, was fitted by making the sum

$$\sum_{A} \sum_{Z(A)} FIY(A,Z) - \sum_{A} 1 = 0$$

and this turned out to be too loose a constraint. The fitting was therefore recast so that

$$\sum_{Z(A)}^{\Sigma} FIY(A,Z) - 1 = 0$$

for each mass number A.

The equations to be solved are then 1, 2, 3 above together with

It was required to minimise

$$\chi^{2} = \frac{\Sigma}{A} \left\{ \frac{\left[\Upsilon(A) - \gamma(A)\right]^{2}}{\sigma^{2}(A)} + \frac{\Sigma}{Z(A)} \left( (FIY(A, Z) - fiy(A, Z))^{2} \cdot fiy(A, Z) \right) \right\}$$

y(A) being the experimental value for the fission yield of chain mass A, and fiy(A,Z) the independent yield of nuclide (A,Z) as calculated from the experimental data, the adjustments being

$$\alpha(A) = Y(A) - y(A)$$
  
$$\beta(A,Z) = FIY(A,Z) - fiy(A,Z)$$

It is then required to solve

$$\frac{\partial \chi^2}{\partial \alpha(A)} + \lambda_1 \frac{\partial \varphi_1}{\partial \alpha(A)} + \lambda_2 \frac{\partial \varphi_2}{\partial \alpha(A)} + \dots + \lambda_{93} \frac{\partial \varphi_{93}}{\partial \alpha(A)} = 0$$

$$\frac{\partial \chi^2}{\partial \beta(A,Z)} + \lambda_1 \frac{\partial \varphi_1}{\partial \beta(A,Z)} + \lambda_2 \frac{\partial \varphi_2}{\partial \beta(A,Z)} + \dots + \lambda_{93} \frac{\partial \varphi_{93}}{\partial \beta(A,Z)} = 0$$

for all A,Z, together with equations  $\emptyset_1$  to  $\emptyset_{93}$ , Y(A) being replaced by (y(A)+ $\alpha$ (A)) and FIY(A,Z) by (fiy(A,Z)+ $\beta$ (A,Z)).

The solution boils down to solving approx. 950 simultaneous equations, the matrix of which is very sparse. Dr. R.K. Reid of CSSD suggested and realised the method of solution.

There remain to be added the further constraints

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This part of the work is now in progress.

Comprehensive review of all fission yield data for the IAEA Fission Product Nuclear Data Panel meeting ; Patten, September 1977 (J.G. Cuninghame (AERE))

Completed and published as AERE-R8753 (1977).

3. Related Compilation Studies

### The CASCADE programme (G. Evangelides (Imperial College) and D.G. Vallis (AWRE)

The computer programme CASCADE<sup>(1)</sup> interprets evaluated decay schemes in terms of radiation data sets and it is the objective of this work to expand and improve the code to provide a data base for input to the programme.

In the course of the work modifications have been made both to the original version of CASCADE and to the variable dimensioned format versions written at Imperial College. The codes' capabilities have been extended to handle proton and neutron emission and to account for any decay of the parent nuclide by spontaneous fission. Some minor modifications are still required and these will be implemented at the same time as changes to the decay type reference number. The latter changes are required to achieve consistency with the ENDF/B IV and V formats. Tobias'<sup>(2)</sup> code for calculating the average energy of  $\beta$ -spectra is being tested and an option for producing output data in ENDF/B format is being written. These will both be sub-routines of CASCADE.

A code has been written in variable dimentioned format to obtain input data from the NSDF/ENSDF compilations<sup>(3)</sup>. The virtue of this format is that it enables the programme to deal with the varying numbers of levels in different decay schemes. In a section of the data on the original magnetic tape from America, columns 1 to 6 have been shifted by one column to the left, column 1 being blank. The first part of the code corrects this and a header section is added to each decay scheme to make access to and handling of the data more easy. Specific decay schemes may be extracted from the data tape or the whole may be converted into CASCADE input format.

THE NDSF compilations have been successfully rewritten and the two files seem to contain about the same amount of information of use to CASCADE but ENSDF also contains about 5 or 6 times as many isotopes with only the description of their adopted levels and without the primary decay data to these levels.

The data of Band et al<sup>(4)</sup> for internal conversion cofficients have been punched onto cards and when checked will be added to the existing data of Hager and Seltzer<sup>(5)</sup>. The Kondurov et al<sup>(6)</sup> data on the life times of nuclear levels have also been punched onto cards and could be used to supplement the NSDF/ENSDF data.

The CASCADE output data are being catalogued by using the IBM SORT/MERGE routines. These routines have been fully set up and a number of codes have been written which will list output data in the order required.

It is intended that the output data will be used with the following programmes: FISPIN-4<sup>(7)</sup> - the Harwell version will be used, its data supplemented by CASCADE output data.

GAMLEG-W<sup>(8)</sup> - this code for the production of multigroup photon transport cross sections is now available on the Harwell computer. It has been compiled and sample problems executed successfully.

ANISN-W<sup>(9)</sup>

solves neutron and photon transport calculations. It has been converted back to IBM format and compiled successfully. Sample programmes are being tested.

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- (6) I.A. Kondurov et al., INDC (CCP)-89/N
- (7) B.L. Richardson, Unpublished Information
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- (9) R.G. Saltesz, R.K. Disney, WANL-PR(IL)-034, V4. (1970)

### DIVISION OF RADIATION SCIENCE AND ACOUSTICS, NATIONAL PHYSICAL LABORATORY

(Superintendent: Dr. W.A. Jennings)

Various methods of measuring neutron flux density are being developed, employed and intercompared as a means of improving the accuracy of the existing standards. Neutrons in the 1 - 1000 eV energy range (E.J. Axton and A.G. Bardell)

No further progress has been made in the provision of a low energy neutron source based on the Van de Graaff accelerator as the machine has not been available for this work.

Measurements to determine the importance of the contribution of neutrons in this energy range to the total dose equivalent in a typical power station location have been completed. The measurements were made using polyethylene spheres with gold foils at the centre. The efficiency matrix for the spheres was determined by Monte Carlo calculations using ENDF/B4 neutron cross-section data, supported by calibrations at a few neutron points. Methods of interpretation of the data are being investigated.

There appear to be relatively more low energy neutrons in the neutron spectrum at the power station site than there are in the '1/E' spectrum of the NPL thermal column.

One of the spheres which was exposed in the 2 keV reactor filtered beam at the Physikalisch-Technische Bundesanstalt (PTB), West Germany, indicated a significantly higher dose-rate than would be expected from the stated fluence.

### Intermediate energy neutrons (J.B., Hunt and R.A. Mercer)

The measurement of the neutron detection efficiency of the NPL standard long counter and of a De Pangher long counter, at 920 keV, 727 keV and 582 keV, using the associated target activity method, and employing the  ${}^{57}$ Fe(p,n) ${}^{57}$ Co reaction, is now completed. The induced activity in twelve different  ${}^{57}$ Fe targets has been redetermined and the analysis completed except for a few minor corrections. The results confirm the previous determination of the long counter efficiencies to better than 3%, based on calibrated radioactive neutron sources with an overall uncertainty of about 3%. The efficiency of a De Pangher long counter has also been measured at 2 keV and 25 keV using the reactor filtered beams available at P.T.B. and the analysis of the data is being completed.

Fast neutron energies (T.B. Ryves, K.J. Zieba and P. Kolkowski)

The iron and copper cross-section measurements have been completed and accepted for publication in Metrologia (July 1978 issue), with the following abstract:-

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'The cross-sections for the reactions  ${}^{56}$ Fe(n,p) ${}^{56}$ Mm,  ${}^{63}$ Cu(n,2n) ${}^{62}$ Cu, and  ${}^{65}$ Cu(n,2n) ${}^{64}$ Cu for neutron energies from 14 to 19 MeV were measured by the activation technique, using a proton recoil telescope to determine the neutron fluence. The uncertainties, ranging from 2 to 6%, were estimated at approximately the 99% confidence level. Less accurate measurements of the  ${}^{54}$ Fe(n,2n) ${}^{53}$ Fe,  ${}^{65}$ Cu(n,p)  ${}^{65}$ Ni and  ${}^{65}$ Cu(n,n $\alpha$ )  ${}^{61}$ Co cross-sections were also made. A detailed account of the experimental procedure is given, and the results are compared with several recent evaluations. No fine structure was observed in the  ${}^{56}$ Fe(n,p)  ${}^{56}$ Mm cross-section over the energy-range from 17.5 to 17.9 MeV.

The half-life of  $^{62}$ Cu was found to be 9.73 ± 0.06 min, where the uncertainty is approximately at the 99% confidence level.

The measured  ${}^{56}$ Fe(n,p) ${}^{56}$ Mn cross-section will be used as a secondary standard at the NPL for determining monoenergetic neutron flux densities from 14 to 19 MeV by the activation technique'.

Further activation cross-sections are now being measured, including  ${}^{14}N(n,2n)$ ,  ${}^{19}F$  (n,2n),  ${}^{27}Al(n,p)$  and  ${}^{27}Al(n,\alpha)$ .

## Neutron source calibrations (E.J. Axton and A.G. Bardell)

Improvements in analytical techniques have demonstrated the presence of some hitherto undetected but significant impurities in the manganese sulphate bath solution. Work so far completed indicates that it may be necessary to re-value upwards by an amount of the order of 0.2% the emission of the neutron sources calibrated with this equipment.

A new international intercomparison of neutron source emission rates based on a circulating source of <sup>252</sup>Cf has been arranged under the auspices of the International Bureau of Weights and Measures (BIPM) to take place in 1978/79. Possible sources of discrepancy revealed in the last intercomparison<sup>(1)</sup> could arise from inaccuracies in <sup>56</sup>Mn counting, and the presence of unsuspected impurities. Precautions are being taken to reduce these possible sources of discrepancy in the next intercomparison.

Differences in the measurement of the neutron escape from the manganese sulphate bath have been found to be due to the presence of neutron-producing impurities, e.g. beryllium, fluorine, within the neutron source itself.

#### Progress in the BIPM international intercomparison of neutron flux density

Iron foils have been sent to PTB and NRC (Canada) as part of the 14 MeV intercomparison based on the  ${}^{56}$ Fe(p,n) ${}^{56}$ Mn reaction. These measurements will formally mark the end of the intercomparison. Transfer method studies are in progress for a new

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intercomparison to begin in 1979 in which it is hoped that 'white-source linacs' will participate.

#### Reference standard for fast neutron dosimetry (V.E. Lewis, E.J. Axton, D.J. Thomas and D.J. Young)

Collimated beams of 14.7 MeV neutrons have been produced. Modifications have been made to the 150 kV accelerator to improve output and reliability. A duoplasmatron ion source is being constructed. A beam line of the Van de Graaff accelerator has been set up specifically for the dosimetry of 5.5 MeV neutron fluences.

Cavity ionisation chambers for mixed field dosimetry have been made and a proton recoil telescope suitable for measurement of kerma in hydrogen completed. The sensitivites to 5.5 and 4.2 MeV neutrons for several types of GM dosemeter have been measured. A dualphosphor thermoluminescence dosimetry system is being developed for measurements made at other centres, with samples sent to NPL by post.

#### Nuclear decay scheme measurements (P. Christmas, D. Smith, M.J. Woods, R.A. Mercer P. Cross and S.P. Brown)

An improved system for the measurement of half-lives has been constructed and is operating satisfactorily. Half-life measurements have been made for  $^{203}$ Pb,  $^{99}$ Mo,  $^{85}$ Sr and  $^{57}$ Co. Work is continuing on the measurement of the  $\gamma$  branching ratio in the decay of  $^{86}$ Rb by absolute counting techniques, while the iron-free,  $\pi$   $\sqrt{2}$  magnetic  $\beta$ -ray spectrometer has been applied to the precise determination of internal conversion coefficients and the  $\gamma$ branching ratio in the decay of  $^{137}$ Cs.

#### W-value measurements (P. Christmas, M. Burke and I. Brearley (Birmingham University)

W-values for protons of energy 700 keV to 3 MeV in gases of interest in neutron dosimetry are to be determined at Birmingham University under an NPL contract, and at NPL. The necessary apparatus has been constructed and measurements are already under way at Birmingham.

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#### PUBLICATIONS

V.E. Lewis and D.J. Young, Phys. Med. Biol. Vol. 22 (1977) 476 'Measurement of the Fast-Neutron Sensitivities of Geiger-Mueller Counter Gamma Dosemeters'

M.J. Rossiter, V.E. Lewis and J.W. Wood. Phys. Med. Biol., Vol. 22, (1977) 731 'The response of Thermoluminescence Dosemeters to Fast (14.7 MeV) and Thermal Neutrons'

#### PHYSICS RESEARCH AND DEVELOPMENT, NATIONAL RADIOLOGICAL PROTECTION BOARD

Group Leaders: B.L. Davies (Applied), J.A. Reissland (Experimental and Theoretical) Neutron facility (C.L. Harvey, E.A. Pook)

The Board has installed a SAMES TD8 accelerator at Harwell. The accelerating potential is 300 kV. Two beam lines are available, a 3 metre line in the straight ahead position and an 8 metre line at an angle of  $30^{\circ}$ . The deuteron beam is deflected into the long arm by a bending magnet. The long arm terminates over a large pit to enable measurements to be made which require low interference from scatter. A tritium target is used to produce 14.6 MeV neutrons (<sup>3</sup>He(d,n)<sup>4</sup>He) and a deuterium target to produce 3 MeV neutrons (<sup>2</sup>H(d,n)<sup>3</sup>He).

Beam currents of 5 mA have been obtained in the  $0^{\circ}$  and  $30^{\circ}$  directions respectively. A tritium target in the  $0^{\circ}$  arm has yielded 7 x  $10^{11}$  neutrons per second and it is anticipated that  $10^{12}$  neutrons will be produced with a fresh target. Using the  $30^{\circ}$  arm, a tritium target should produce about 3 x  $10^{11}$  neutrons per second over the pit. The deuterium target is now being fitted to the  $30^{\circ}$  beam line and about 5 x  $10^{10}$  neutrons per second are expected.

#### Neutron detection (A.G. Sherwin, J.C.H. Miles and J.B. O'Hagan)

The section has developed various detector systems for the measurement and characterisation of the mixed neutron and gamma-ray fields produced by the SAMES accelerator and other neutron sources.

A neutron spectrometer system for measurements in the energy range 10 keV to 20 MeV consists of an organic liquid (NE 213) scintillator and a set of four hydrogen-filled spherical proportional counters, each detector covering a particular part of the energy range. The neutron spectrum is unfolded from the measured data by an iterative computer program developed within the department. This program required a detector response matrix which has been calculated using the 'Monte Carlo' method. A set of activation and fission foils is also used for spectral measurements.

A pair of ionisation chambers have been designed and constructed for the accurate determination of the absorbed dose components in mixed fields. One chamber is hydrogenous and the other non-hydrogeneous, which means they have considerably differing neutron sensitivities, while photon response is similar. A conventional subtraction technique is employed for dose component separation.

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The section has also developed a new method for mixed field analysis (1,2,3). This relies on the application of statistical analysis to the fluctuating ionisation current induced by the mixed field in a single ionisation chamber.

Fission foils have been utilised as secondary standards for the measurement of fast neutron fluence  $^{(4,5)}$ . The foil of fissile material is placed in contact with polycarbonate film, and the nuclear fragments produced by neutron induced fission cause damage tracks in the plastic which are subsequently revealed by chemical etching. The neutron fluence is determined by counting the fission fragment tracks using a microscope.

- A.G. Sherwin, in Proc. 2nd Symp. on Neutron Dosimetry in Biology and Medicine, Munich (EUR 5273), (1974)
- (2) A.G. Sherwin, in Monograph on Basic Physical Data for use in Neutron Dosimetry (EUR 5629), (1976).
- (3) A.G. Sherwin, in Proc. 3rd Symp. on Neutron Dosimetry in Biology and Medicine, Munich, (1977)
- (4) J.C.H. Miles and A.G. Sherwin, In Monograph on Basic Physical Data for use in Neutron Dosimetry (EUR 5629), (1976)
- (5) J.C.H. Miles and A.G. Sherwin, Nuclear Instruments and Methods, 146, (1977) 503

#### Neutron shielding data (D.G. Jones)

Calculations have been made to determine the neutron energy spectrum at different points inside a steel shield irradiated with 14.6 MeV neutrons from the facility described above. The calculations were performed using the neutron transport code DOT3.5<sup>(1)</sup> and the multigroup cross section data taken from EURLIBI data set<sup>(2)</sup>. The work formed part of the Intercomparison Benchmark Shielding Project.

The figures (1-4) show the neutron spectral distribution at the four positions were measurements are being made.

 F.R. Mynatt, F.J. Muchkenthaler and P.N. Stevens, 'Development of two Dimensional discrete ordinates transport theory for radiation shielding' CTC - INF - 952, (1969) ESIS Newsletter No. 12, (1975)





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#### Neutron cross-sections (P.J. Dimbylow)

The advent of high energy neutron radiotherapy has created a need for an evaluated set of neutron cross-section data for the main constituents of tissue, i.e. H, C, N and O, for energies up to 50 MeV. The experimental data between 20 and 50 MeV are so few (except for H) that nuclear model calculations have to be employed to produce the required neutron cross-sections. The following computational strategy has been adopted. The total and elastic cross-sections are calculated by an optical model fit to the experimental total cross-section data. The non-elastic cross-sections produced can then be used to normalise reaction cross-sections calculated on the basis of the complete statistical model.

The optical model program, NUPTIC, has been developed. The Bjorklund-Fernbach potential, which has a maximum of 9 independent parameters, is used. However, because of the paucity of experimental data, 7 of these parameters were kept constant with energy and a fit to the data was performed by varying the real and imaginary well depths. It was found that a simple linear variation provided a good fit to the data, the real depth decreasing and the imaginary depth increasing. The graph (fig. 5) shows an optical model fit (curve) to the experimental total cross-section data<sup>(1,2,4,5)</sup> for oxygen.

A computer program which considers the emission of up to three particles has been written, based on the complete statistical model with a Fermi gas level density function. Inverse capture cross-sections were taken from the work of Dostrowsky et al.<sup>(3)</sup>. Averaged values for pairing energies were taken from Vonach and Hilke<sup>(6)</sup>. The level density parameter, "a" is initially set to  $^{A}/8$  for each residual nucleus and then is adjusted if necessary to fit experimental reaction cross-section data. Calculations based on this program are almost complete, although at energies around 50 MeV it may be necessary to consider the emission of up to 6 particles for oxygen and nitrogen.

 M. Auman, F.P. Brady, J.A. Jungerman, W.J. Knox, M.R. McGie, and T.C. Montgomery, Phys. Rev., <u>C5</u>, (1972) 1

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- (5) J.M. Peterson, A. Bratenahl and J.P. Stoering, Phys. Rev. 120, (1960) 521
- (6) H. Vonach and M. Hilke, Nucl. Phys., A127, (1969) 289





Fig. 5 Optical model fit (curve) to the total cross section of  $^{16}$  O

#### UNIVERSITY OF BIRMINGHAM, DEPARTMENT OF PHYSICS

(Professor of Applied Nuclear Science, J. Walker)

# Experimental Neutronics Studies for CTR Blankets (M.C. Scott, N. Evans, J. Jowett, J. Perkins, B.Y. Underwood)

The first stage of the experimental programme involving measurements of absolute neutron spectra in a large assembly of LiF using the Birmingham Radiation Centre's 3 MV Dynamitron accelerator has now been completed, and the results are being assessed to determine what the emphasis should be in the next stage. The work can be considered in three sections. In addition, feasibility studies related to time-dependent measurements have been undertaken and are discussed in the forth section below.

#### (a) Neutron yield determination from the (D,T) source

In order to make the measurements absolute (i.e. neutron flux per source neutron) the target yield during the spectrum measurements has to be known. The associated particle technique was used in two configurations: for low current measurements a very small assembly (~ 2.5 cm diameter) was made in which the incident deuteron beam passed through the centre of an annular surface barrier detector, the alpha particle being detected in the backward direction. For higher currents the alpha particle was also detected in the backward direction, but by a (3 mm diameter) surface barrier detector placed off-axis to the beam. In order to check that the calculated relationship between alpha count rate and neutron yield was correct, measurements of the absolute neutron yield were made using a continuous flow vanadium bath assembly<sup>(1)</sup>, for which the leakage and charged particle losses for a 14 MeV source were calculated using ANISN<sup>(2)</sup>. The measurements were made at a range of incident deuteron energies, from 250 to 500 keV, and over this energy range the two techniques agreed to within 10% (fig. 1). It is possible that the error assigned for the charged particle cross sections used in ANISN are too low, in which case this systematic difference could be within the uncertainties involved. One effect which was not quantified was the count rate from the decay of  ${}^{16}N$  following the  ${}^{16}O(n,p){}^{16}N$  reaction, <sup>16</sup>N having a 7s half life. This is currently being investigated: any allowance which has to be made will improve the agreement.



Fig. 1 Values of the neutron to alpha ratio for a miniature tritium target

# (b) Neutron spectrum measurements in the energy range from 40 keV to 2.5 MeV using proton recoil proportional counters

Measurements were made at a number of radial positions in the 1.25m diameter sphere of LiF using two counters, one filled with hydrogen to 2.5 atmospheres and the other with hydrogen and argon to a total of 10 atmospheres. Two parameter accumulation (rise time and pulse height) was used in both cases, proton recoil events being stripped off using Gaussian fitting functions. It was particularly interesting that the argon recoil events could also be identified quite clearly<sup>(2)</sup>. Figure 2(a) (b) shows two sets of results, the comparison being with calculations using ANISN with the ENDF/BIV data sets for fluorine and lithium. There was very good agreement at the outside of the sphere, but there was increasing disagreement towards the source. Implications of these discrepancies from the data adjustment point of view are still under consideration.

#### (c) <u>Neutron spectrum measurements in the energy range from 500 keV to 16 MeV using a</u> <u>miniature NE213 scintillation counter</u>

Several different glass cells containing ~1.5cm<sup>3</sup> of NE213 have been constructed, and a range of studies made to investigate the effect of different reflecting surfaces and of the geometry on the detector response. For a given configuration, a significant difference was found in the measured resolution between different scintillator samples, and this could not be accounted for. In general, however, the results were in agreement with the photon transport predictions which form part of the programme.



Fig. 2(a) Absolute proportional counter measurements of the neutron flux at 55.4cm in a 1.25m diameter lithium fluoride assembly







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Fig. 3 Calculated values of the pulse shape dependence on particle energy in NE213

The use of two parameter accumulation (pulse shape and energy) has made it possible to distinguish pulses from  $\gamma$ -rays, recoil protons, and  $\alpha$ -particles from the  ${}^{12}C(n,\alpha)$  and and  ${}^{12}C(n,3\alpha)$  reactions. In practice, the two parameter trace of the  $\alpha$ -particles crosses that from the recoil protons (see figure 3), a phenomenon which has now been accounted for theoretically taking into account the energy dependence of dE/dx and the light output for the two particles.

Using this detector, absolute measurements have been made in the LiF assembly at a number of points: the results of one such measurement are shown in fig. 4, where they are compared with an ANISN calculation using ENDF/BIV data. In order to represent the source



Fig. 4 Scintillation Counter Measurements of the absolute neutron flux in a 1.25m diameter lithium fluoride assembly

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properly, more recent work uses the Monte Carlo code MORSE with much finer lethargy steps and allowance for an anisotropic source: however, the results shown here assume an isotropic source with the neutrons assigned to one lethargy group. The effect of the target angular energy dependence shows clearly in the spectra measured at different angles.

Because the neutron flux in the system is very anisotropic, the choice of appropriate response functions was investigated theoretically using the 05S Monte Carlo code with modified input routines to allow neutron incidence at any angle. It was found that the efficiency per unit flux was virtually independent of angle, but the effect of angle of incidence on the response function shape is still being studied; the results so far, for our own system, indicate that the differences were small, i.e. within the statistical errors of the present calculations.

#### (d) <u>Time-dependent sensitivity studies</u>

A computer-based investigation was carried out to assess whether slowing-down time measurements in the LiF sphere might be used for cross-section evaluation. Experiments of this type using a pulsed neutron source have already been performed in this laboratory<sup>(3)</sup>, utilizing the time-dependent gamma-ray production rate from inelastic indicators to provide information on the time development of the neutron energy spectrum in the 100 nanosecond time range. In the present case it was envisaged that the miniature liquid scintillator (having intrinsic fast timing capability) would provide a more efficient and direct way of measuring the time-dependent neutron spectrum.

The study made use of the MORSE multigroup time-dependent Monte Carlo computer programme<sup>(4)</sup>. The procedure was to make physically significant types of perturbation to elements on the ENDFE IV cross-section data at about the 10% level, and to observe the effect on the time development of either the flux spectrum itself or of the response of a liquid scintillator. This can be compared with the effect of such perturbations on steady-state parameters. Straightforward application of a Monte-Carlo programme for perturbation analysis is normally unsatisfactory, since the effects of small perturbations on parameters of interest may be hidden in statistical fluctuations of the Monte Carlo estimates from run to run. To overcome this problem a method of correlated sampling was used<sup>(5)</sup> in which the estimates for different perturbations were based on the same set of Monte Carlo histories, and the dominant effect of statistical fluctuations thereby removed.

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The investigation revealed that the perturbations of cross-sections close to the source energy have the most significant effect on steady state and time-dependent spectra: this agrees with SWANLAKE sensitivity studies for the steady-state spectrum. However, the main effect on the time-dependent spectra is a change in overall magnitude rather than a change in shape, indicating that such time-dependent measurements are unlikely to provide much extra information on high-energy cross-sections. It should be noted, however, that the range of perturbations tried was by no means exhaustive.

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#### PHYSICS DEPARTMENT, UNIVERSITY OF EDINBURGH

(Department Head: Prof. W. Cochran)

Polarization and differential cross-section for the elastic scattering of 16 MeV neutrons (R.B. Galloway and F. McNeil Watson) [Relevant to request numbers: 169-72, 174, 559]

Measurements are continuing of the angular dependence of polarization and of the differential cross-section over the angular range  $20^{\circ}-160^{\circ}$  for the elastic scattering of 16.1 MeV neutrons by Fe, Cu, I, W, Hg and Pb. The 16.1 MeV polarized neutron beam is obtained from the  ${}^{3}$ H(d,n)<sup>4</sup>He reaction using IBIS. The present set of polarization measurements are compared in fig. 1 with optical model calculations. The solid curves are for calculations with the parameter set of Rosen<sup>(1)</sup>, the dashed curves for the parameters of Becchetti and Greenlees<sup>(2)</sup> and the dotted curve for Pb is from the parameters of Fu and Perey<sup>(3)</sup>. In all cases around  $20^{\circ}$  there is a clear difference between the measurements and the calculations. This feature will be investigated further once the differential cross-section measurements are completed.

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- (2) F.D. Becchetti and G.W. Greenlees, Phys. Rev. 182 (1969) 1190
- (3) C.Y. Fu and F.G. Perey, Atomic Data and Nuclear Data Tables, 16 (1975) 409

The polarization of 14 MeV neutrons due to elastic scattering (Amena Begum and  $R_{\circ}B_{\circ}$  Galloway)

In view of the significant discrepancy between the measured and calculated polarization values for the elastic scattering of 16 MeV neutrons through  $\sim 20^{\circ}$ , reported above, measurements on 14 MeV neutrons are being made. Since 14 MeV neutrons from the  ${}^{3}\text{H}(d,n)^{4}\text{He}$  reaction induced by deuterons of a few hundred keV are unpolarized, a double scattering experiment is being carried out. The measured asymmetry in the second scattering gives the square of the polarization. A preliminary measurement on  $20^{\circ}$  scattering by Cu give  $|P|^{2} = 0.3 + 0.1$  which may be compared with 0.25 + 0.05 from the 16 MeV data.

An improved version of the experimental system is under construction and further measurements should be made in the next few months.



Fig. 1 Polarisation measurements. The points are the present measurements, the curves are optical model calculations with the parameter set of Rosen(1) (solid curve) and Bechetti and Greenlees<sup>(2)</sup> (dotted curve). The dotted curve for Pb is from the parameters of Fu and Perey(3)

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#### Polarization and differential cross-section for the elastic scattering of 2.9 MeV neutrons (Amenga Begum and R.B. Galloway)

Measurements made recently in this laboratory of the polarization and differential cross-section for the elastic scattering of 2.9 MeV neutrons by Fe, Cu, I, Hg and Pb<sup>(1)</sup> are being augmented by measurements on W, Tl, Pb, Bi and U. Relevant optical model and Hauser Feshbach calculations are also in progress.

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 R.B. Galloway and A. Waheed, Proc. 4th Int. Symp. on Polarization Phenomena in Nuclear Reactions (eds. W. Gruebler and V. Konig, Birkhauser Verlag) Basel, (1976) 605

The <sup>9</sup>Be(d,n)<sup>10</sup>B reaction as a source of polarized neutrons (B.S. Bains and R.B. Galloway)

The  ${}^{9}\text{Be}(d,n){}^{10}\text{B}$  reaction is one of the more prolific neutron sources available with low energy deuterons and for that reason it's suitability as a source of polarized neutrons for elastic scattering measurements has been investigated. Polarization measurement was by elastic scattering by <sup>4</sup>He. The neutron group leading to the ground state of  ${}^{10}\text{B}$  was found to provide a neutron beam with a polarization of 0.35 + 0.06 at a reaction angle of  ${}^{45}\text{}^{\circ}$  to a 400 keV deuteron beam. The discrimination required to reject the lower energy neutrons leading to higher excited states of  ${}^{10}\text{B}$  is similar to that required to reject inelastically scattered neutrons in typical elastic scattering measurements.

A full description of the investigation has been published.<sup>(1)</sup>

(1) B.S. Bains and R.B. Galloway, Nucl. Instr. and Meth. 143 (1977) 295

## Neutron polarization in the reaction $7_{Li(d,n)}^{8}$ Be (R.B. Galloway and A.M. Ghazarian)

Continuing the investigation of neutron producing reactions accessible with a low energy accelerator which may serve as a source of polarized neutrons for elastic scattering measurements, the  ${}^{7}$ Li(d,n) ${}^{8}$ Be reaction is now being studied. Elastic scattering by  ${}^{4}$ He is being employed as polarization analyzer. A preliminary measurement on the ground state neutron group emitted at 55° to a 450 keV deuteron beam indicates a polarization of less than 0.1.

# The polarization of neutrons from the ${}^{2}$ H(d,n) ${}^{3}$ He reaction for deuteron energies from 35 to 275 keV (A.M. Alsoraya and R.B. Galloway)

An account of this investigation, described in last year's progress report, has been published in Nuclear Physics A280 (1977) 61.

#### A gaseous electric-field-enhanced scintillation detector for angular distribution measurements of fission fragments (D.E. Cumpstey and D.G. Vass)

Development has continued of the xenon gas scintillation detector with electric field enhancement of the light emission described in a previous edition of this report.<sup>(1)</sup>

When a fission fragment, from a <sup>252</sup>Cf source mounted on the cathode plane in such a detector, is stopped in the xenon gas, the electron cloud formed along its track subsequently drifts under the influence of the applied electric field towards the anode plane. Excitation of the gas occurs and photons emitted during the de-excitation process are detected by a photo-multiplier. Also to a first order approximation the range of the fission fragment depends only on its initial kinetic energy and not on its atomic mass and charge numbers.<sup>(2)</sup>

The emission rate of fragments from the source with energies in the range E to E+dE into the solid angle  $2\pi d\cos\theta$  about the direction  $\theta$  to the axis of the detector is  $2\pi R$  $(E,\cos\theta)dEd\cos\theta$ . For a thin, uniform source where the emission is isotropic,  $R(E,\cos\theta)$  is of course independent of  $\theta$  and may be written as  $\Phi(E)$ . Fragments of energy E emitted at angle  $\theta$  generate scintillation pulses of amplitude V and of fall time T determined by the duration of the collection of the primary electrons at the anode. We have observed that VaE for  $\theta \stackrel{<}{\sim} 86^{\circ}$ , and so using the appropriate energy calibration we may measure the distribution N(E,T) over a wide range of values, where N(E,T)dEdT =  $\tau 2\pi R(E, \cos\theta) dEd\cos\theta$ ,  $\tau$  being the measuring time. Or simply, N(E,T)dT =  $a\Phi(E)d\cos\theta$ , **a** being a constant.

Noting that the minimum and maximum values,  $T_{MIN}$  and  $T_{MAX}$  of the fall time of the scintillation pulses occur for values of  $\cos\theta$  equal to 0 and 1 respectively, the angle  $\theta$  at which a fragment of energy E is emitted may be deduced from the measurement of the fall time T of its scintillation pulse by exploiting the relationship for the yield(E,T  $\rightarrow T_{MAX}$ ) of fragments with fall times between T and  $T_{MAX}$  to determine the " $\cos\theta$ -T" calibration curve, namely,

$$\cos = 1 - \{y(E,T \rightarrow T_{MAX}) / y(E,T_{MIN} \rightarrow T_{MAX})\}$$

Here  $y(E,T \rightarrow T_{MAX}) =$ 

$$\int^{T} MAX N(E,T) dT$$

=  $a\Phi(E)$  {1 -  $\cos \theta$ }, with 0 <  $\theta$  <  $\frac{\pi}{2}$ 

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The energy distributions of fragments from the <sup>252</sup>Cf source for the range of  $\cos \theta$ values indicated, and determined by the above method, are presented in figure 1. Ideally, the distributions should all be identical, but there is a spoiling of the energy resolution as  $\theta$  increases to  $\frac{\pi}{2}$ . This we attribute to energy absorption in the source of the fragments emitted at grazing angles to its surface.

- D.E. Cumpstey and D.G. Vass, U.K. Nuclear Data Progress Report, U.K.N.D.C. (76)
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Fig. 2 Energy distributions of fission fragments emitted at different angles from a <sup>252</sup>Cf source using the techniques described in the text

#### KELVIN LABORATORY, UNIVERSITY OF GLASGOW

(Director: Professor R.O. Owens)

## 1. Inelastic neutron scattering from <sup>89</sup>Y (G.I. Crawford, S.J. Hall, J.D. Kellie, P.W. Martin, G. Cleland

Following our measurement of the neutron total cross-section of  $^{89}$ Y (UKNDC(77)P86, p53) we are continuing our study of that nucleus by an experiment to detect the de-excitation gamma-rays from  $^{89}$ Y irradiated with fast neutrons. A 12% efficient Ge-Li detector was used with an annular target containing approximately 400gm of Yttrium; these were placed 50m from the neutron-producing target of tantalum which was exposed to 60 ma pulses of 100 MeV electrons, each 3ns wide, at a repetition rate of 600 Hz. The neutron output of the accelerator was continuously monitored by detectors placed in adjacent beam lines of our multi-angle facility. This monitoring system enabled us conveniently to check the target-out background (which was negligibly small) and the incident neutron energy spectrum (for which a small liquid scintillator proton recoil detector was used.

Fig. 1 shows the total  $\gamma$ -ray spectrum (i.e. the pulse height spectrum of all events in the GeLi detector which arrived between 0.8 and 4.0 µs after a beam pulse) recorded during the experiment. The seven numbered peaks in fig. 1 are tentatively identified with the seven numbered transitions in the partial level scheme for <sup>89</sup>Y which is shown in fig. 2. Confirmation of the assignment will come from an examination of the time-offlight spectra which are in coincidence with the relevant photopeaks. Fig. 3 shows one such time spectrum, that in coincidence with photopeak 1, which exhibits a very sharp cut-off around channel 350. The corresponding energy spectrum (fig. 4) shows that this corresponds to a sharp rise in yield of the  $\gamma$ -ray just above 1.5 MeV, exactly as expected.

Analysis of this data is in progress.

### 2. Low energy delayed neutrons from <sup>95</sup> Rb (G.I. Crawford, S.J. Hall, J.D. Kellie)

In experiments with a <sup>3</sup>He spectrometer K.L. Kratz et al. (c.f. I.L.L. Annual Report 1977) have found very many peaks in the energy range from 13 keV to over 1 MeV in the energy spectrum of delayed neutrons which follow the decay of <sup>95</sup>Rb. Since their claimed neutron groups at 13.6, 25 and 48 keV were unresolved in their original spectrum and were only revealed by an unfolding procedure we have undertaken a further study of this decay using the time-of-flight technique. A thin NE 102 plastic scintillator detected the  $\beta$ -particles while an 11.0 cm diameter by 1.27 cm thick NE912 lithium glass scintillator was used as a neutron detector. Several flight paths in the range 35 to 55cm were used.

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Fig. 1 Total gamma ray spectrum from  $^{89}$  Y(n,n'Y)



Fig. 2 Partial level scheme for  $^{89}$ Y



Fig. 3 Time of flight spectrum for neutrons observed in coincidence with photopeak 1



Fig. 4 Energy spectrum corresponding to the time spectrum of fig. 3

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On the basis of the time resolution measured with  $\gamma$ -rays we estimate our resolution to be 0.2 keV at 12 keV and 0.5 keV at 25 keV neutron energy. We also used a 12.5cm by 5.08cm NE213 liquid scintillator detector to study the high energy part of the spectrum, at flight paths of 1 and 1.5m. So far only the lowest energy region has been finally analysed. Fig. 5 shows the sum of 4 separate runs taken at 3 different flight paths. We conclude

- (1) that there is a prominent group of delayed neutrons of energy 13.7  $\pm$  0.2 keV, whose width is 0.9  $\pm$  0.2 keV
- (2) there are other significant peaks at 11.2 and 25.5 keV, each of width 2 keV The efficiencies of our detectors were measured relative to the <sup>252</sup>Cf fission spectrum and we hope to have available shortly a corrected yield curve covering the whole energy range with Li glass detector and from about 300 keV upwards with the liquid scintillator.

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Fig. 5 Energy spectrum of delayed neutrons from <sup>95</sup>Rb observed by neutron time of flight spectroscopy

3. Delayed neutrons from <sup>17</sup>N (G.I. Crawford, S.J. Hall, J.D. Kellie, G. Cleland)

The beta decay of  ${}^{17}$ N is followed almost 100% of the time by neutron emission, the neutron energy spectrum consisting of 3 distinct peaks at about 385, 1165 and 1175 keV. If a convenient way can be found to produce  ${}^{17}$ N this will provide a useful source for neutron detector calibration. There are also physics reasons for studying this decay. For instance, a recent paper (Ohm et al, Nuclear Physics A274 (1976) p45) has suggested that a fourth weak group of neutrons exists at 850 keV, and also that the previously reported widths of the neutron groups had been underestimated.

We have carried out a preliminary experiment which has established the feasibility of using the  ${}^{18}_{0}(\gamma,p){}^{17}_{N}$  reaction with a 1g sample of water enriched to over 90% in  ${}^{18}_{0}$ . Only a very short (0.6m) flight path was used but the resolution and statistics achieved suggested that the 3 main neutron groups did indeed have widths as suggested by 0hm et al.

This work is continuing. A full scale run will be undertaken shortly with a flight path of 4m, in order to provide good resolution at the highest energy.

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