1212

UNCLASSIFIED

UKNDC(82)P105
NEANDC(E)232 Vol.8
INDC(UK)-36/LN

UKNDC(82)P105
NEANDC(E)232 Vol.8
INDC(UK)-36/LN

United Kingdom Atomic Energy Authority

HARWELL

UK Nuclear Data Progress Report for the Period January - December 1981

Editor: E.W. Lees
Nuclear Physics Division
AERE Harwell, Oxfordshire
June 1982

UNCLASSIFIED

UNCLASSIFIED

UKNDC(82)P105
NEANDC(E)232 VOL.8
INDC(UK)-36/LN

U.K. NUCLEAR DATA PROGRESS REPORT FOR THE PERIOD JANUARY - DECEMBER 1981

Editor: E.W. Lees

Nuclear Physics Division
AERE Harwell

June 1982

HL82/1736

CONTENTS

*For a key, see the Introduction to Chapter 1 on page 23.

*For a key, see the Introduction to Chapter 1 on page 23.

PREFACE

This report is prepared at the request of the United Kingdom Nuclear Data Committee and covers the period from January to December, 1981.

Nuclear data are presented by laboratory. There are contributions this year from the Harwell and Winfrith laboratories of the UKAEA, the National Physical Laboratory, the National Radiological Protection Board, the Birmingham Radiation Centre, the University of Birmingham and the University of Edinburgh.

This report includes work from various collaborations between laboratories of Harwell, Dounreay, Winfrith, Windscale, MOD Aldermaston, Imperial College and Manchester University. Contributions on "Chemical Nuclear Data" are gathered by the Chemical Nuclear Data Committee and grouped under that heading.

Contributions to the report on nuclear data topics are welcome from all sources and we extend an invitation to researchers in other laboratories of industry, government, the universities and polytechnics to use this channel of communication.

Where the work is clearly relevant to requests in WRENDA 81/82 (INDC(SEC)-78/URSF) request numbers are given after the title of the contribution.

NUCLEAR DATA FORUM LECTURES

The fifteenth Nuclear Data Forum was postponed from its traditional December date by severe weather. The rescheduled meeting took place at Harwell on 28th January 1982 with 65 participants. The themes this year were thermal reactor data needs and fission product decay heat. The invited talks were given by Dr. J.R. Askew (review of thermal reactor data needs) and Mr. M.F. James (review of calculations and measurements of decay heat); unfortunately, only the latter is reproduced below. Eight of the thirteen contributed talks gave direct support to the main speakers, while the others covered aspects of data requirements for radiation dosimetry calculations, unresolved resonance region in ^{238}U, delayed neutron energy spectra and ν for spontaneous fission.

FISSION PRODUCT DECAY HEAT FROM ^{235}U AND ^{239}Pu - A BRIEF SURVEY OF THE EXPERIMENTAL AND THEORETICAL DATA

M.F. James

AEE, Winfrith

Lecture given at the Nuclear Data Forum, AERE Harwell

1. Introduction

Decay heat from fission products has been of great importance since the early days of reactor design, but in the last few years work on this topic has burgeoned throughout the world: an excellent review has been published by Tobias[1]. There are three main reasons for this recent increase in effort. First, several very careful measurements of total decay heat or of its beta and gamma components have been made in a number of different laboratories. Secondly, computer power has increased sufficiently for rapid summation calculations to be feasible; and thirdly, better data for the libraries used by these summation calculations are available from measurements of half-lives, fission yields and decay schemes of a large fraction of all fission products.

However, it is the aim of this paper to emphasise that there are still unresolved discrepancies between calculation and measurement and between different measurements, and to suggest that the recommended uncertainties in predicted decay heat should reflect these discrepancies.

2. Current UK Recommendations

A detailed study in 1978 of calculation and measurements of decay heat made the following recommendations for use in the United Kingdom:-

(i) Predictions of decay heat should be made with a summation program such as FISPIN[2] or FISP[3], using the UK Fission Product Decay Data Library I (UKFPDD1)[4] and the set of fission yields evaluated by Crouch[5] and designated Crouch 2 or C2.

(ii) for ^{235}U, the uncertainties in the total decay heat following an infinite irradiation are:
7% for cooling times less than 200 seconds,
5% at longer cooling times
(both 1 standard deviation).

(iii) for ^{239}Pu, the uncertainty in total decay heat following an infinite irradiation is 10% (1 standard deviation).

The uncertainties for ^{235}U were chosen:-

(a) from a study of U.S.[6,7] and French[8] estimates of uncertainties in

calculations, taking into account the larger uncertainties in Crouch's fission yields compared with those of Meek and Rider used in U.S. calculations[9].

(b) after analysis of the calculated decay heat obtained using different data libraries

and

(c) from a comparison of calculated and measured decay heat.

At the time of this 1978 study the only measurements available of ^{239}Pu total decay heat were those of Fiche[10], of Gunst[11], and of Dickens et al at ORNL[12]; in addition preliminary results had been announced from measurements at LASL.

It appeared from these that there were larger discrepancies than with ^{235}U, and hence the recommended uncertainty was made greater; this also reflected the greater uncertainties in fission yields for ^{239}Pu.

Since 1978 there have been a number of important developments. Data libraries have considerably improved, largely due to the availability of more and better experimental information on yields, half-lives and decay schemes, especially for short lived neutron-rich nuclides. In the UK a revised version of the fission product decay data library UKFPDD2[13] has been produced, and in addition Crouch has revised his fission yields by putting extra physical constraints in his fitting[14]; this new yield set is designated Crouch 3I or C3I*

Secondly more measurements have been reported especially for ^{239}Pu; in particular we comment here on those on total decay heat from ORNL[12], LASL[15] and IRT[16], and on those on beta heat at Winfrith[22].

Finally the FISPIN summation program has been extended[17] to allow computation of the sensitivities of decay heat to changes in yields and decay data.

3. The Significance of the Ratio of ^{239}Pu to ^{235}U Decay Heat

Besides comparing measurements and calculations of ^{239}Pu and ^{235}U decay heat, it is also useful to compare calculated and experimental values of the ratio of ^{239}Pu to ^{235}U decay heat because there is considerable correlation between the data for the two nuclides (and of course for any other fissile nuclides). That this is so for calculations was pointed out

---

*The "I" indicates that this set contains yields for isomeric states, which was not the case for the original Crouch 2 set.

by Trapp and Spinrad[18]. The yields are of course different for the two nuclides, but the fission product decay data (half-lives, energies and branching ratios) are common, although some chains are much more significant for one nuclide than for the other.

For measurements, one expects that some of the systematic errors of a particular method (e.g. in determining the number of fissions and in detector efficiency) are common to each nuclide and will thus tend to cancel out in the ratio.

Of major importance is a combined study of the ^{235}U data, the ^{239}Pu data, and the data for the ratio $^{239}Pu/^{235}U$ by means of which we hope to decide between the two hypotheses:-

(i) data for ^{235}U are good, but those for ^{239}Pu are relatively poor

or

(ii) data for the two nuclides are equally in error, and the apparent good fit between calculation and measurements for ^{235}U is fortuitous.

Until this decision has been made, it is wise not to reduce the present recommended uncertainties.

4. Comparison of Measurements and Calculations

The decay heat measurements studied have used a variety of irradiation times (I) and cooling times (t), making immediate intercomparison difficult. To aid comparison several techniques can be used and we discuss each in turn.

4.1 Intercomparison Using C/E Values

Each measurement can be compared with the results of a summation calculation for the appropriate I and t, and then values of the ratio C/E of calculation to experiment can be compared.

Figs. 1-6 show C/E ratios for several experiments. The first two graphs show C/E for both ^{235}U and ^{239}Pu total decay heat for the measurements by Dickens et al at ORNL[19,12] (Fig. 1), and for the measurements by Yarnell and Bendt at LASL[20,15] (Fig. 2). In each case the curves for the two nuclides are very similar in shape but C/E for ^{235}U is about 6-8% greater than that for ^{239}Pu. The results are summarised in the following table:-

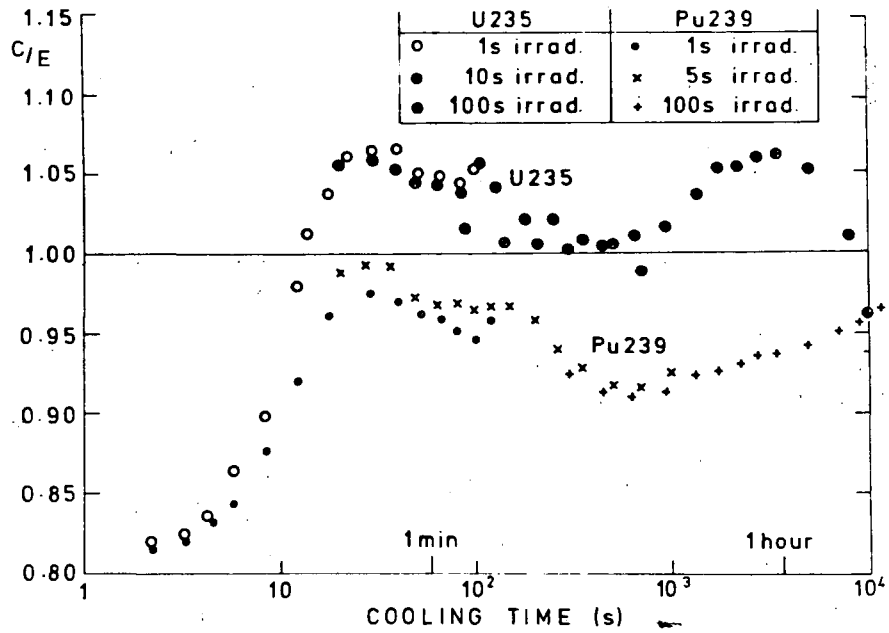

Fig. 1. Comparison of UKFPDD-1/C2 calculations of the total decay heat with the ORNL measurements.

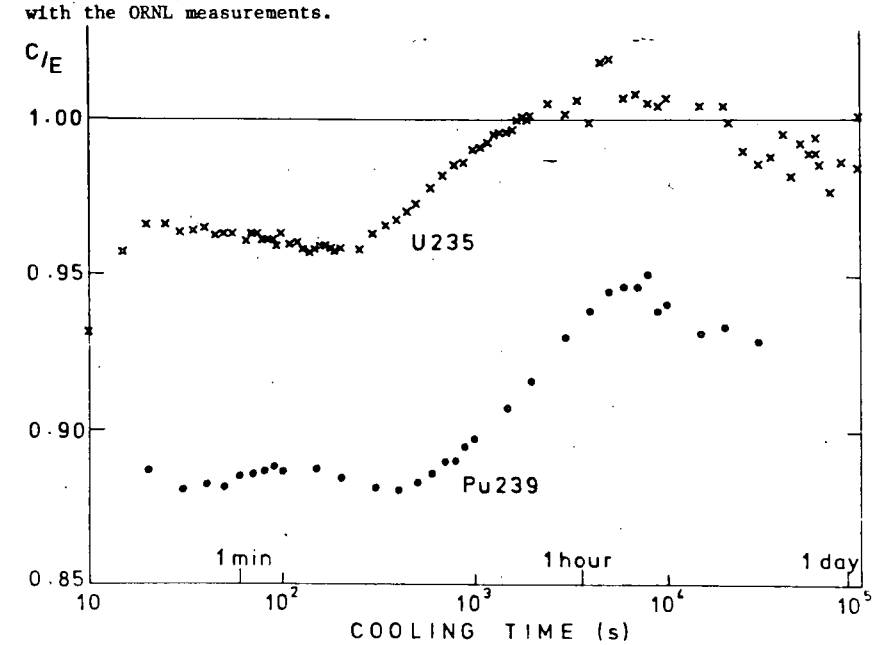

Fig. 2. Comparison of UKFPDD-1/C3I calculations of the total decay heat for ^{235}U (x) and ^{239}Pu (•) after 2 x 10^4s irradiation with LASL measurements.

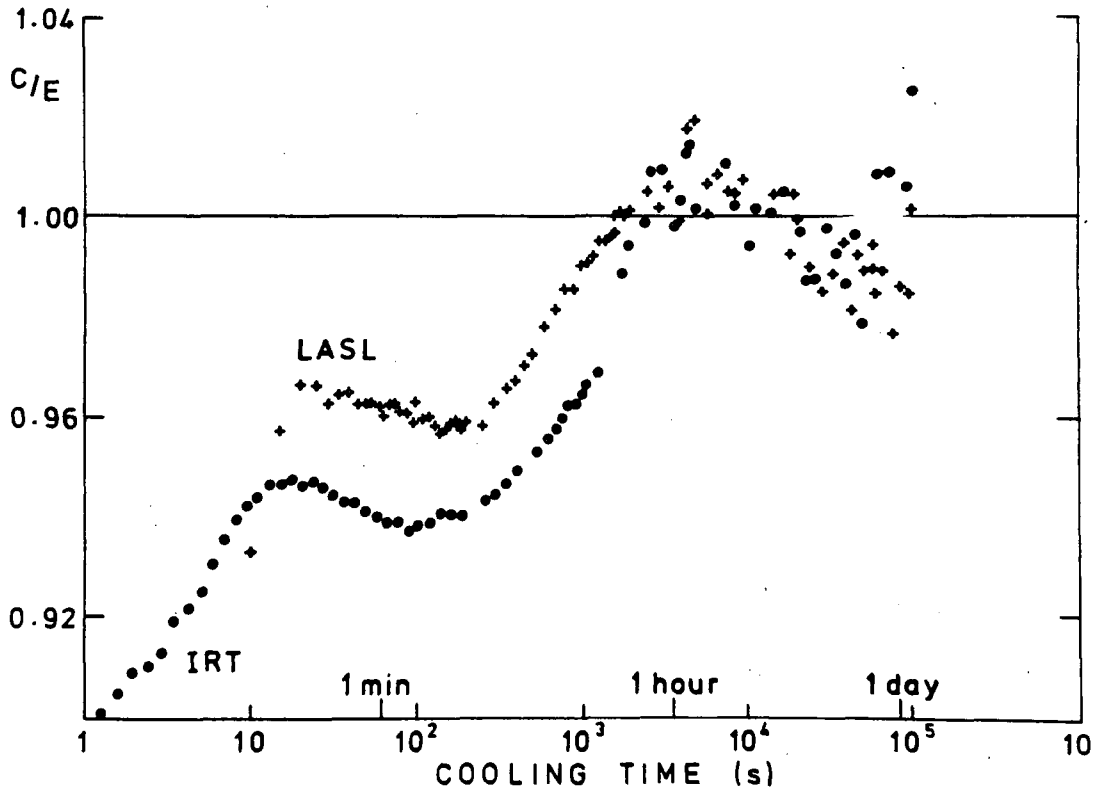

Fig. 3. Comparison of UKFPDD-1/C3I calculations of the total decay heat for ^{235}U after 2 x 10^4s irradiation with the LASL measurements (+) and the IRT measurements (●).

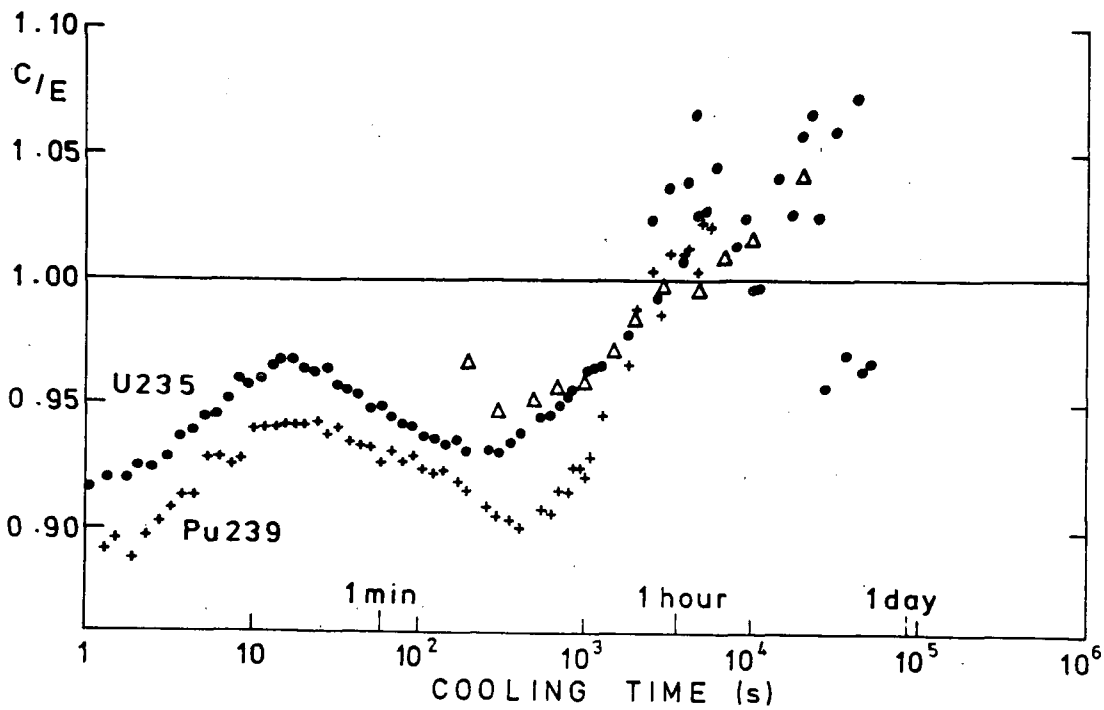

Fig. 4. Comparison of UKFPDD-1/C3I calculations of the total decay heat after 10^3s irradiation with IRT measurements on ^{239}Pu (+) and for ^{235}U (●) also shown for ^{235}U (Δ) are the measurements of ref. (21).

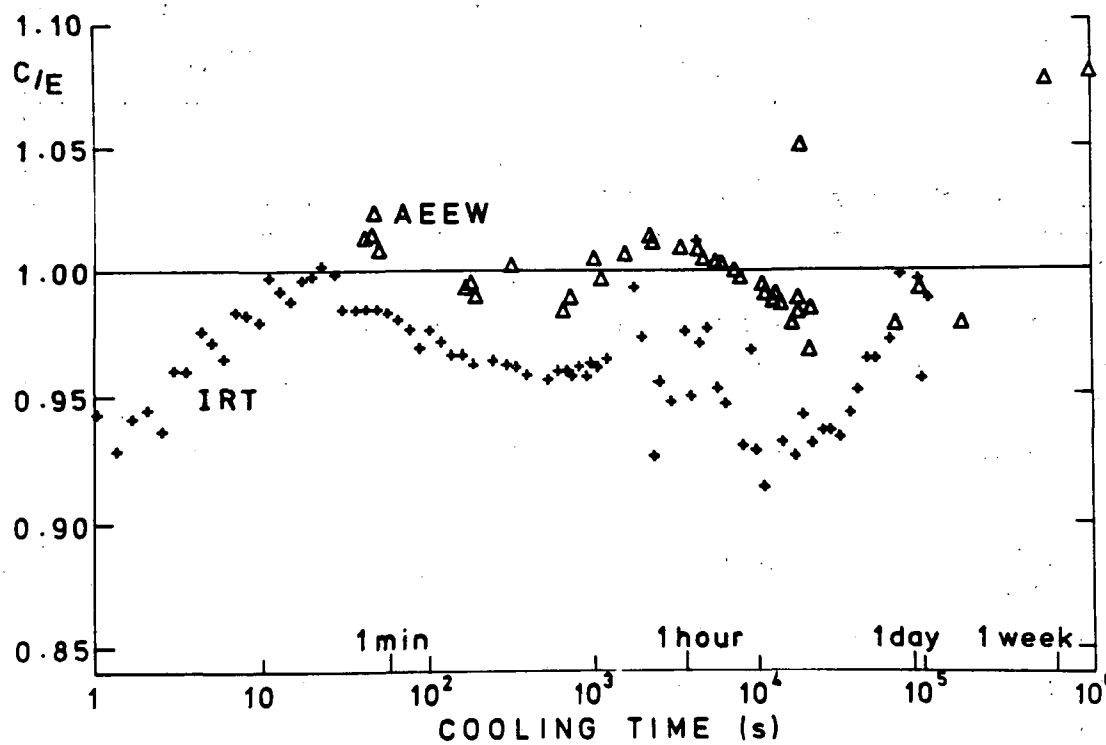

Fig. 5. Comparison of UKFPDD-1/C3I calculations of the beta heating for ^{235}U with IRT measurements (+) after 8.64 x 10^4s irradiation and also with AEEW measurements (Δ) after 10^5s.

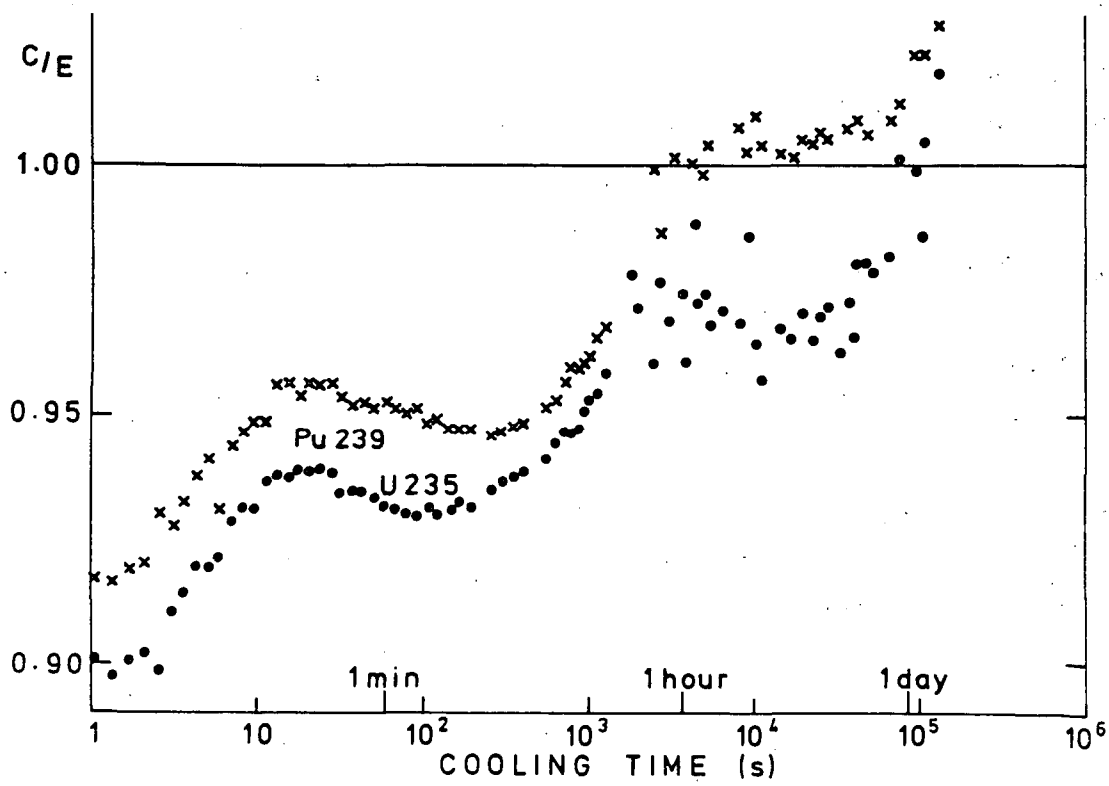

Fig. 6. Comparison of UKFPDD-1/C3I calculations of the total decay heat after 24 hours irradiation with IRT measurements on ^{235}U (●) and ^{239}Pu (x).

Experiment	C/E (^{235}U)	C/E (^{239}Pu)
ORNL (I = 1,10 and 100s)	1.00 to 1.06; C/E ≈1.03	0.91 to 0.99; C/E ≈0.95
LASL (I = 20,000 s)	0.96 to 1.02; C/E ≈0.99	0.88 to 0.95; C/E ≈0.92
IRT (I = 20,000 s)	0.94 to 1.00; C/E ≈0.97	

Fig. 3 compares C/E values for LASL and IRT measurements[16] on ^{235}U, after an irradiation of 20,000 secs; the table also summarises the latter. For 10 sec < t < 1000 sec, the LASL measurements are systematically ~2% below the IRT values. At longer cooling times agreement is better but the scatter in both sets of points implies an increase in random error to ~±1%.

Fig. 4 shows C/E for measurements with I = 1000 secs; for ^{235}U, there is good agreement between IRT and French measurements[21] except at about 200-300 secs, where systematic errors in the latter may be appreciable. C/E for ^{239}Pu is about 2-3% lower than for ^{235}U.

Fig. 5 compares C/E for beta heating measurements on ^{235}U at Winfrith[22] and IRT[16]; the irradiation times although not identical were very similar (about 1 day). Again a similarity in shape can be seen with a systematic difference of about 3%; the Winfrith values are close to 1.00.

Fig. 6 shows results for IRT measurements of total decay heat after an irradiation of 1 day. The notable features here are the much poorer values of C/E and the reversal of the situation with respect to other measurements in that ^{239}Pu decay heat is calculated more accurately than ^{235}U decay heat (by about 2%).

4.2 Comparison of Measurements by Synthesis of Different Irradiations

The decay power t seconds after an irradiation of 1 fission per second lasting for I seconds is:-

$$M(I,t) = \int_{t}^{I+t} m(x)dx$$

where m(x) is the decay power x seconds after a single fission. (The small effect of neutron capture is ignored.)

Therefore,

$$M(nI,t) = \sum_{K=0}^{n-1} M(I,t + KI)$$

Consequently values for one irradiation can be built up by summing data from a shorter irradiation if the smaller value of I is a factor of the larger, and if the cooling times after the shorter irradiation extend to more than the longer irradiation.

To overcome experimental fluctuations and to allow interpolation at different cooling times, a sufficiently smooth curve must be fitted to the results of the shorter irradiation; a least squares cubic spline program written at Harwell was used for this.

Some results are shown in Fig. 7 comparing LASL measurements (I = 20,000 s) with values computed from short irradiation ORNL measurements, and in Fig. 8, comparing IRT measurements (I = 1000 sec) with ORNL values.

It is apparent that there are considerable differences between the different measurements in that LASL results are up to 8% higher than ORNL, and IRT and ORNL results differ by 8-12%.

4.3 Reduction of Measurements to Burst Functions or to Infinite Irradiation

If $I \ll t$, $M(I,t) \approx Im(t + I/2)$, and so the "burst function", m(t), giving the decay power after a single fission may be estimated.

Conversely the decay heat after an infinite irradiation;

$$M(t) = M(\infty,t)$$

may be calculated from

$$M(t) = M(I,t) + M(\infty,I + t)$$

where for $I \gg t$, the second term on the right is a small correction that can be estimated from summation calculations.

These methods of reporting results have been used in several measurements particularly those at ORNL and in France. However, they are only "self-contained" without relying heavily on other results for the extreme cases of $I \ll t$ to $I \gg t$.

5. (^{239}Pu/^{235}U) Ratios

As explained in Section 3 it is valuable to consider the ratio of ^{239}Pu to ^{235}U decay heats. For convenience we write:-

$$R(I,t) = \frac{M(I,t) \text{ for } ^{239}\text{Pu}}{M(I,t) \text{ for } ^{235}\text{U}}$$

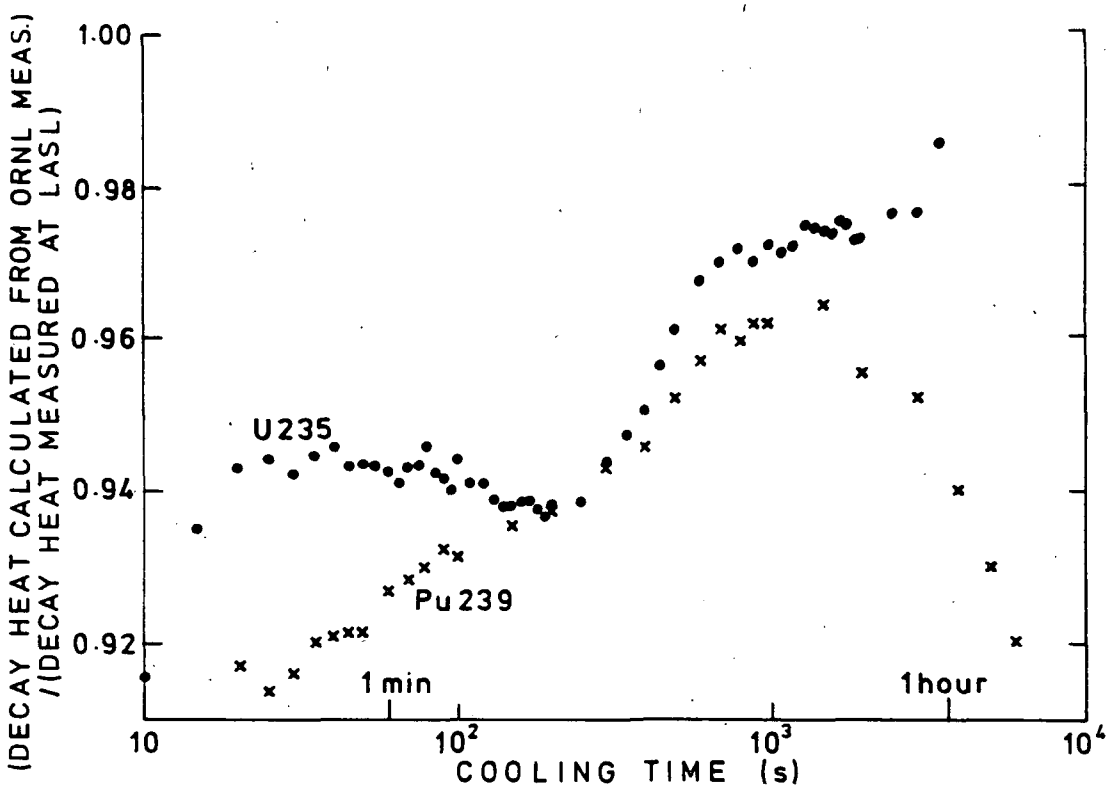

Fig. 7. Comparison of the total decay heat computed from short irradiation measurements at ORNL and compared to measurements at LASL after 2 x 10^4s irradiation for ^{235}U (●) and ^{239}Pu (x).

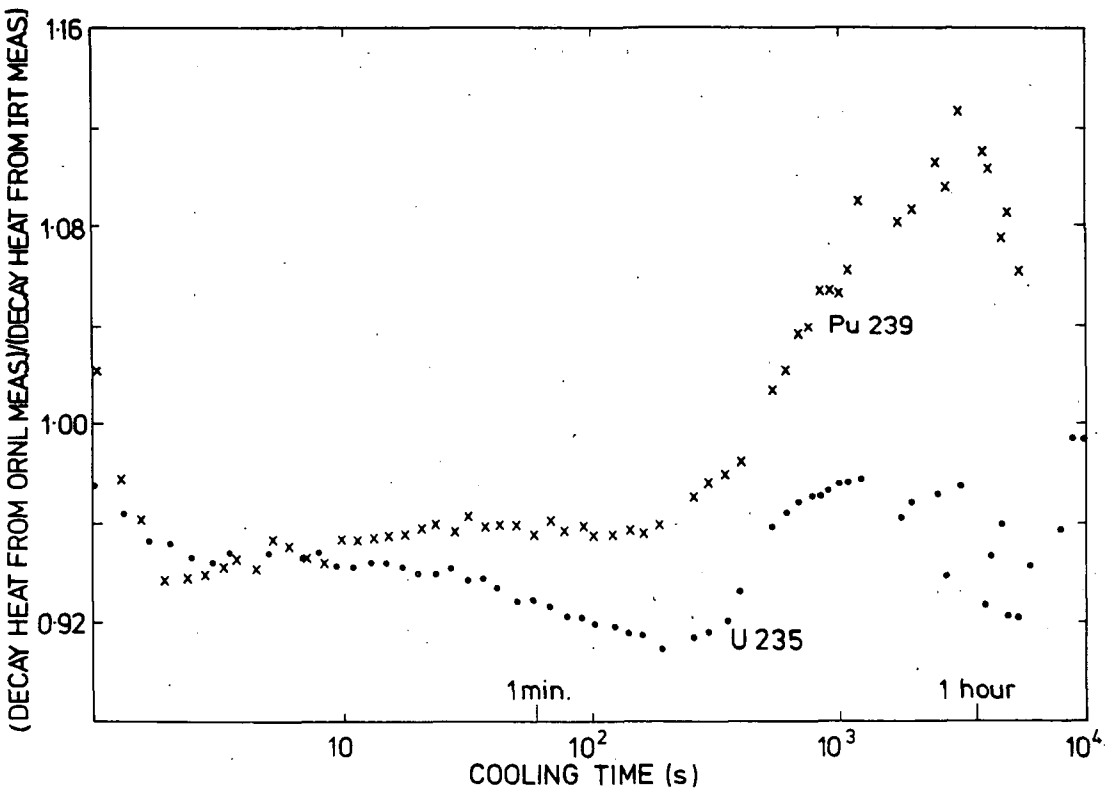

Fig. 8. As for Fig. 7 but for an irradiation time of 10^3s and IRT measurements.

Fig. 9 shows results for I = 20,000 secs comparing (a) measurements from LASL[20], (b) values derived from ORNL measurements[12,19] by the method described in Section 4.2; and (c) calculations using UKFPDD-1 and C3I.

The LASL and ORNL measurements agree remarkably well particularly for cooling times greater than 100 secs. It will be remembered that Fig. 7 showed a considerable difference (of up to 8%) between the two sets of measurements for each nuclide separately: hence we may deduce that most of the systematic error in the measurements at either laboratory is the same for each nuclide, as suggested in Section 3.

The two experimental curves have very similar shapes to the calculated values but it may be seen that:-

$$\frac{\text{Calculated } R(20,000,t)}{\text{Measured } R(20,000,t)} \approx 0.92$$

Fig. 10 compares the ratio of decay powers for single fissions from ^{239}Pu and ^{235}U:

$$r(t) = \frac{m(t) \text{ for } ^{239}Pu}{m(t) \text{ for } ^{235}U}$$

The experimental values shown are those derived from measurements at ORNL[12,19] and at the CEA[21,10]. Calculated values of r(t) were obtained using the C3I yield set with UKFPDD-1 and UKFPDD-2; the change in decay data makes only slight differences except at short times.

The two sets of experimental results agree well especially for $100s < t < 10^4 s$, but there is again a discrepancy between calculation and experiment:-

$$\frac{\text{Calculated } r(t)}{\text{Measured } r(t)} \approx 0.91 \text{ for } 100s < t < 10^4 s$$

It is interesting to speculate whether the resolution of this discrepancy in r(t) would resolve the discrepancy in the 20,0000 s irradiation values plotted in Fig. 9.

6. Summary and Conclusions

(1) There are systematic differences between decay heat measurements from different laboratories. Some may be due to errors in estimating the number of fissions, but as there is dependence on cooling time apparent in the discrepancies, part is presumably due to errors in estimating the efficiency of the detector or calorimeter (which would be dependent on beta or gamma spectrum and hence on cooling time).

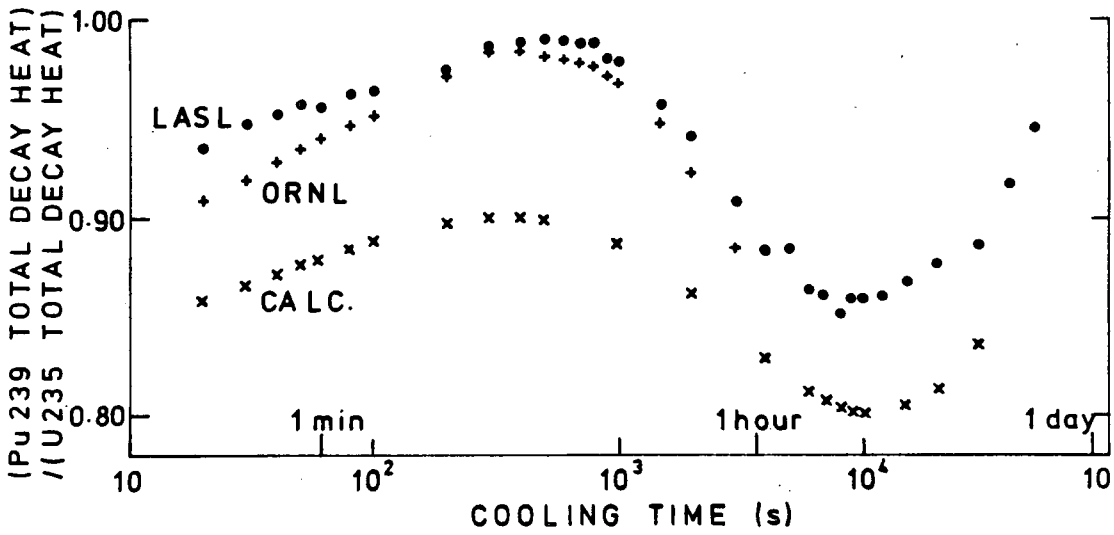

Fig. 9. Ratio of total decay heat for $^{239}Pu/^{235}U$ after 2 x 10^4s irradiation; data are (●) lasl measurement, (+) derived from ORNL and (x) calculated using UKFPDD-1/C3I.

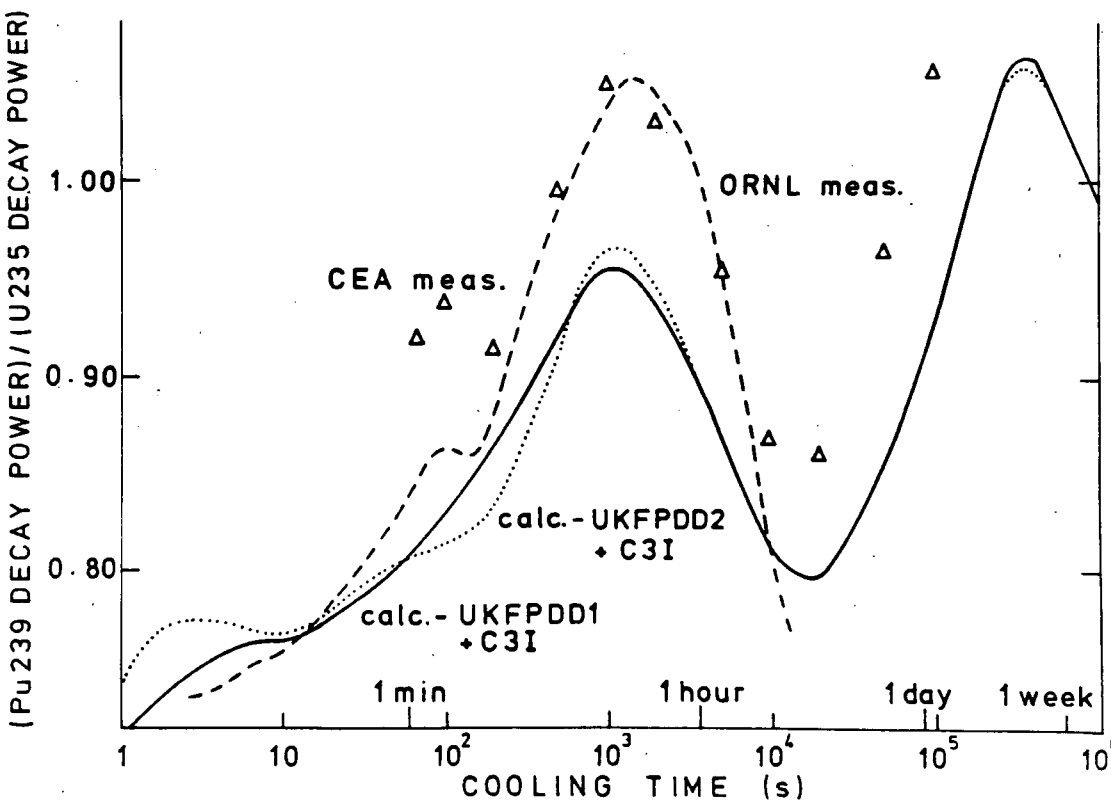

Fig. 10. Ratio of total decay powers after 1 fission for $^{239}Pu/^{235}U$; data are (Δ) CEA, (- - -) ORNL, (——) calculated using UKFPDD-1/C3I and (.....) calculated using UKFPDD-2/C3I.

(2) Most of these differences, at least for some of the sets of measurements disappear if the ratio of ^{239}Pu to ^{235}U decay heat is considered, implying that most of the systematic errors are independent of fissile nuclide, or if they are dependent on fissile nuclide are the same for each laboratory.

(3) Calculated values for ^{235}U fall roughly between the different sets of measurements, but for ^{239}Pu the calculation generally underestimates decay heat.

(4) The calculated values for the ratio of ^{239}Pu to ^{235}U decay heat are about 8% or more below the experimental values.

(5) No discussion has been given in this paper of possible errors in the decay data or fission yield libraries, the sensitivity program FISPIN-SENS is still being developed and results so far are only preliminary. It does not appear easy however to suggest errors of sufficient magnitude to explain the above discrepancies. One area of uncertainty must be the fission yields for ^{239}Pu. The extra physical constraint of making yields of complementary elements equal that was introduced by Crouch to obtain the Crouch 3 set gave some improvement but only of about 2% at the cooling times of interest. There may be some errors in the data in the mass chains A ~105 - 110 which are significant for ^{239}Pu but not for ^{235}U fission, but they would need to be quite large to resolve the discrepancy.

(6) Until the discrepancy is resolved, either by discovering systematic errors in the measurements, or errors in the yields or decay data, or perhaps by a combination of the two, there does not seem to be any justification for changing the current recommendation on uncertainties.

(1) A. Tobias. Prog. in Nucl. Energy 5(1), pp 1-93 (1980).

(2) R.F. Burstall. UKAEA Report ND-R-328(R) (1979).

(3) A. Tobias. CEGB Report RD/B/N4303 (1978).

(4) A. Tobias. CEGB Report RD/B/N4053 (1977).

(5) E.A.C. Crouch. At. Data and Nucl. Data Tables 19, 417 (1977).

(6) R.E. Schenter, F. Schmittroth and T.R. England. IAEA-213, Vol. 2, p. 677 (1978).

(7) M.A. Bjerke, J.S. Holm, M.R. Shay and B.I. Spinrad, Nuclear Safety 18, 596 (1977).

(8) C. Devilliers et al. NEACRP L163.

(9) J.G. Cuninghame. IAEA-213, Vol. 1, p. 351 (1978).

(10) C. Fiche, F. Durfreche and A.M. Monnier. "Calorimetric Measurements of the Total Power Released by the Thermal Fission Products of ^{233}U and ^{239}Pu". CEA Internal Report (1976).

(11) S.B. Gunst, D.E. Conway and J.C. Connor. Nucl. Sci. Eng. 56, 241 (1975).

(12) J.K. Dickens, J.F. Emery, T.A. Love, J.W. McConnell, K.J. Northcutt, R.W. Peelie and H. Weaver. ORNL/NUREG-34 (1978).

(13) A. Tobias and B.S.J. Davies. CEGB Report RD/B/N4942 (1980).

(14) E.A.C. Crouch. To be published.

(15) J.L. Yarnell and P.J. Bendt. LA-7452-MS, NUREG/CR-0349 (1978).

(16) S.J. Friesenhahn and N.A. Lurie. IRT 0034-004 (1977).

(17) M.F. James. To be published.

(18) T.J. Trapp and B.I. Spinrad. OSU-NE-7801 (1978).

(19) J.K. Dickens, J.F. Emery, T.A. Love, J.W. McConnell, K.J. Northcutt, R.W. Peelie and H. Weaver. ORNL/NUREG-14 (1977).

(20) J.L. Yarnell and P.J. Bendt. LA-NUREG-6713 (1977).

(21) M. Lott, G. Lhiaubet, F. Dufreche and R. de Tourreil, Journ. Nucl. Energy 27, 597 (1973).

(22) M.F. Murphy, W.H. Taylor, D.W. Sweet and M.R. March. UKAEA Report AEEW-R 1212 (1979).

Element S	Element A	Quantity	Type	Energy Min.	Energy Max.	Documentation Ref.	Documentation Vol	Documentation Page	Documentation Date	Lab	Comments
B	10	N xsec	Expt-Prog	Fast	-	UKNDC	P105	62	4/82	HAR	Crouch + check of xsec data in PFR spectra.
Al	27	N alpha	"	1.4 07	-	"	"	36	"	"	Cookson + angular and energy distribution of alphas.
V	Nat	"	"	"	-	"	"	"	"	"	" " "
Ni	Nat	"	"	"	-	"	"	"	"	"	" " "
Ge	68	Half-life	"	-	-	"	"	73	"	NPL	Christmas + To be published $T_{\frac{1}{2}}$ = 270.82 ± 0.27 days.
Nb	93	N, inelast.	"	-	-	"	"	35	"	HAR	Gayther + initial measurements of isomer production.
Nb	93	"	"	Fast	-	"	"	68	"	WIN	Taylor + part of the Nb dosimetry intercomparison.
Eu	152	Half-life	"	-	-	"	"	73	"	NPL	Christmas + To be published.
Eu	154	"	"	-	-	"	"	"	"	"	" " "
U	235	Fiss.Spec	"	Fast	-	"	"	75	"	BIR	Walker + To be published.
U	235	Fiss.Yield	"	"	-	"	"	53	"	HAR	Cunningham + Fission yields for different fast n spectra.
U	235	Decay Heat	Eval	-	-	"	"	8	"	WIN	James + survey of experimental and theoretical data on decay heat.
U	235	N xsec	Expt-Prog	Fast	-	"	"	62	"	HAR	Crouch + check of xsec data in PFR spectra.
U	238	Total xsec	"	1.0 03	1.0 04	"	"	26	"	"	Bee + analysis of thick sample transmission data.
U	238	N xsec	"	"	-	"	"	62	"	"	Crouch + check of xsec data in PFR spectra.
U	238	N gamma	"	1.0 -02	1.0 01	"	"	25	"	"	Moxon + R. matrix shape analysis of data.
U	238	Fiss.Yield	"	Fast	-	"	"	53	"	"	Cunningham + fission yields for different fast n spectra.

Element S	Element A	Quantity	Type	Energy Min.	Energy Max.	Documentation Ref.	Documentation Vol	Documentation Page	Documentation Date	Lab	Comments
Np	237	Half-life	Expt-	-	-	UKNDC	P105	63	4/82	HAR	Glover + planned 1982.
Pu	239	Fiss.spec	"	Fast	-	"	"	75	"	BIR	Walker + To be published.
Pu	239	"	"	"	-	"	"	53	"	HAR	Cuningham + fission yields for different fast n spectra.
Pu	239	Decay heat	Eval	-	-	"	"	8	"	WIN	James + survey of experimental and theoretical data on decay heat.
Pu	239	Half-life	"	-	-	"	"	63	"	HAR	Glover + to be published.
Pu	240	N xsec	Expt-Prog.	Fast	-	"	"	62	"	HAR	Crouch + check of xsec data in PFR spectra.
Pu	241	"	"	"	-	"	"	62	"	"	" " "
Pu	242	"	"	"	-	"	"	62	"	"	" " "
Am	241	"	"	"	-	"	"	62	"	"	" " "
Am	241	N gamma	"	Fast	-	"	"	62	"	"	Anstey + production of ^{242}Cm in Zebra cores.
Am	241	N gamma	"	"	-	"	"	63	"	"	Glover + To be published.
Am	243	"	"	"	-	"	"	63	"	"	" " "
Am	243	N gamma	"	Fast	-	"	"	62	"	"	Anstey + production of ^{244}Cm in Zebra cores.
Am	243	N xsec	"	"	-	"	"	62	"	"	Crouch + check of xsec data in PFR spectra.
Many		N alpha	Eval	1.4 07	-	"	"	72	"	NPL	Hayes + simultaneous evaluation of reaction xsec for several elements.
Many		N elast.	Expt-	3.0 06	-	"	"	76	"	EDI	Annand + angular dependence of polarisation and differential scattering 22 angles.

1. NUCLEAR PHYSICS DIVISION, AERE, HARWELL

(Division head: Dr. J.E. Lynn)

Introduction

Nuclear data measurements in Nuclear Physics Division are diverse and are performed on a variety of sources. Individual research items are labelled with a single letter indicating on which accelerator the experiments were performed. These labels are as follows:

Cockcroft-Walton Generator	A
3 MV pulsed Van de Graaff Generator IBIS	B
6 MV Van de Graaff Generator	C
14 MeV Tandem Generator	D
Electron Linac	E
Variable Energy Cyclotron	G
Synchrocyclotron (now defunct)	H

In the contents pages there is a cross reference to the accelerator where a measurement was made (when this applies).

The material for this contribution is taken from the chapter on Nuclear Data and Technology for Nuclear Power in the 1981 Nuclear Physics Division Progress Report AERE PR/NP29.

1. NUCLEAR DATA AND TECHNOLOGY FOR NUCLEAR POWER

1.1E The new machine for the electron linac laboratory (J.E. Lynn, M.S. Coates, B.P. Clear, J. Down, R.A.J. Riddle and P.W. Swinden)

The machine was accepted provisionally (PR/NP 28, p.8) from the manufacturers in January 1981. Following some electron beam transport difficulties associated with the earth's magnetic field, the Condensed Matter Cell, which is intended primarily for materials physics studies, has become available for experimental runs. It proved necessary to use a combination of current-carrying wires and metal screening to reduce the effects of this field to acceptable levels over the 40 m of electron beam line involved. The target now has had scheduled use for several weeks at mean electron powers of ~20 kW ((~300 joules/pulse, 70 p.p.s.). For a short period the maximum expected beam power of 45 kW was achieved (~300 joules/pulse, 150 p.p.s.). However before this power level is used routinely some alterations will have to be made to some of the collimators and beam scrapers in the electron beam line. At present ~10% of the beam from the linac is lost during transport because of beam clipping and as a result activity is induced in components which leads to difficulties with maintenance. The problem can be remedied by increasing aperture dimensions.

Comparable success has not been achieved in commissioning the experimental cells for nuclear physics use. On the Neutron Booster line it was found that only 50% of the electron beam could be transmitted through the magnet elements of the 90° bend. It has been established that the dipole magnet pole pieces must be re-machined and additional quadrupole elements added to the line before any improvement can be made. This work is underway at present. On the Low Energy line the main problem has been vacuum leaks in magnet box welds. To permit precise magnet field monitoring, these components are made of 310/S24 stainless steel because of its low magnetic permeability. The material is difficult to weld, particularly for complex joins, and microcracks tend to develop. These can grow to large scale cracks during the normal pressure cycles undergone in operational use. New components with simpler welds are being manufactured. Meanwhile low power beam commissioning continues using the old components sealed in a temporary fashion.

No trials have yet been made on the Fast Neutron Cell target because of the difficulties outlined above, but the work will be started soon.

The delays in bringing the installation to a fully operational state have led to a postponement of the work that the manufacturer has to do before the machine is accepted finally.

The klystron harmonic problem (PR/NP 28 p.8) has not been resolved but no further operational difficulties have been experienced. A different type of klystron, manufactured by Litton, has been ordered and its harmonic behaviour will be compared with the Thomson-CSF tubes. The Litton klystron (30 MW peak, 70 kW mean) has a higher power rating that the Thomson-CSF valve (20 MW peak, 40 kW mean) and this is expected to lead to operational advantages. The efficiency of the electron beam injection process on the linac should increase, and the new valve should have a much longer useful life than the Thomson-CSF valve because it is not run close to its maximum rating.

1.2E Measurement of the neutron capture cross section of ^{238}U in the energy range 0.01 to 10 eV (M.C. Moxon and J.E. Jolly)

(Relevant to request numbers 1246 and 1247)

A measurement of the neutron capture cross section of ^{238}U from 0.01 to 10.0 eV was carried out on the 45 MeV electron linac, shortly before it closed down to make way for the 136 MeV machine. The measurement aimed at an accuracy of ±3%. The early attempts at analysis were unsuccessful, partly because of the low signal to background ratio and partly because of inadequacies in the computer code used to correct for multiple neutron interactions in the uranium oxide samples employed. Recently we have re-examined the data using an improved version of the computer code REFIT (PR/NP 26, p.23), which is a least-squares fitting program for neutron resonance analysis. Background can be treated as a variable parameter and multiple neutron interactions can be calculated. The analysis shows that the neutron capture cross section below an energy of a few eV can be represented by a set of recommended parameters using a multilevel formalism. The results have been reported[1] but unfortunately the accuracy requirement for the data is not met. New measurements are planned on the 136 MeV electron linac, where improved signal-to-background conditions are expected. It is hoped also that more suitable samples will be available. Multiple interaction effects are smaller in metallic samples than oxide ones but it has not been possible so far to obtain metal with the desired degree of purity. Trace elements, notably osmium and samarium, introduced during sample preparation contributed significantly to the

observed capture cross section[1] in the samples examined.

---

(1) The Neutron Capture Cross section of ^{238}U from 0.01 to 10 eV. M.C. Moxon and J.E. Jolly, Proc. IAEA Consultants Meeting on Uranium and Plutonium Isotope Resonance Parameters, Vienna (1981) to be published.

1.3 Experimental studies of Doppler broadening in uranium dioxide (N.J. Bee (Imperial College), M.G. Sowerby and M. Bailey)

The work to refurbish the 42 m flight path of the Neutron Booster target (described in PR/NP 28, p.15) is nearly completed and it should now be possible to extend the measurements described in AERE-R 8961. The detector and associated electronics have been extensively modified and the new equipment has been found to work satisfactorily with neutrons from the Condensed Matter Cell target. It is expected that test runs and measurements of the transmission of cold $^{238}UO_2$ can be made shortly after the Neutron Booster comes into operation. Heated sample measurements will then follow.

1.4 Analysis of ^{238}U time-of-flight data in the unresolved energy region (N.J. Bee (Imperial College) and M. Bailey)

This work is reported in detail in ref. 4. It has been shown that cross sections calculated from average transmission values have some interesting properties, apart from depending only slightly on the experimental resolution function. They exhibit a scaling property in the variable $n/\sqrt{\theta}$ where n is the sample thickness and θ is the absolute temperature, and local s-wave strength functions can be extracted from their dependence on this variable. In this way, it has been found possible to isolate contributions of s-wave resonances from other resonances by analysing thick sample transmission measurements as a function of sample thickness or temperature. Previously strength functions could only be obtained as a sum over resonances with different orbital angular momenta, where infinitely dilute cross sections only were used. Fig. 1.1 shows an estimate for the local s-wave strength function for ^{238}U from 1 to 12 keV based on data taken in refs. 1 and 2. The results compare favourably with calculations based on resolved resonances (see ref. 3).

Further work is planned to extend this analysis to cover other data that are available, such as capture data, possibly taking explicitly into account some of the stronger resonances that have been observed in the very high resolution measurements at Oak Ridge[5].

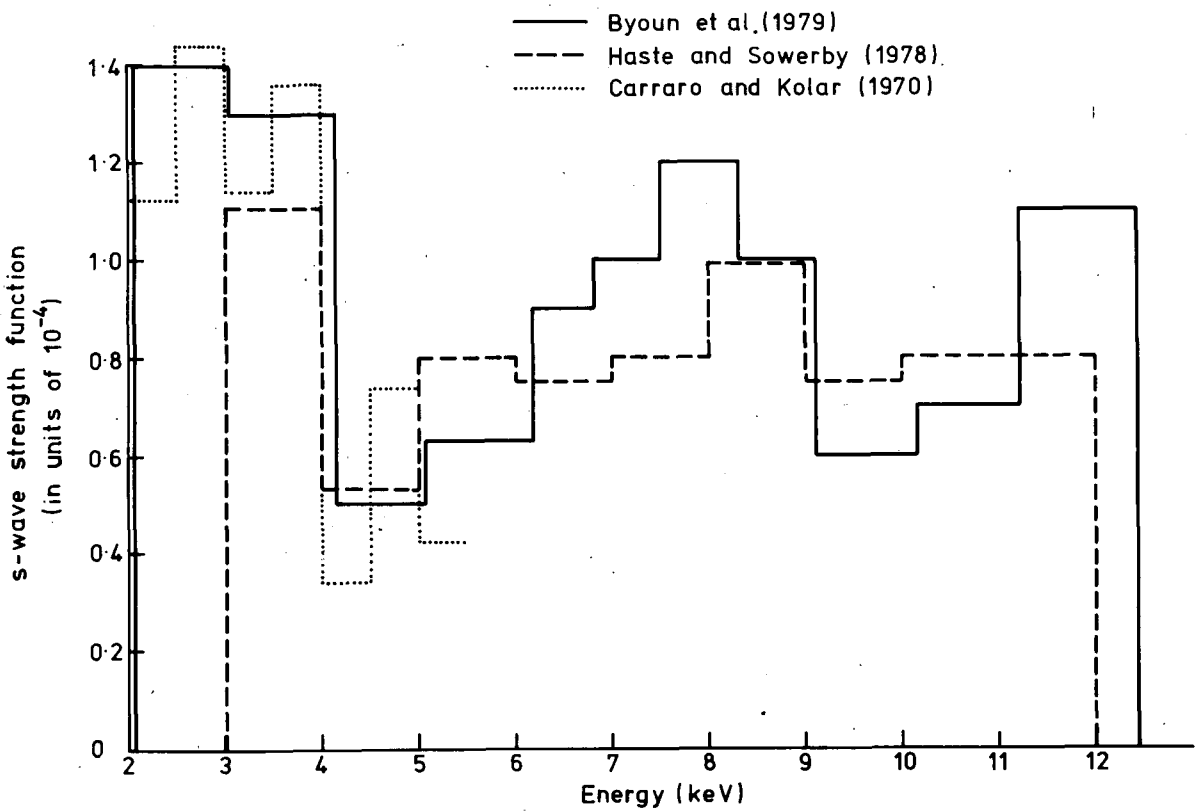

Fig. 1.1 The local s-wave strength function of ^{238}U from 3 to 12 keV, calculated using the techniques of this paper, from data taken by Byoun et al[1] and Haste and Sowerby[2]. A prediction from resolved resonance parameters by Carraro and Kolar[3] is also shown

---

(1) T.Y. Byoun, R.C. Block and T. Semler (1972). Proc. National Topical Meeting on New Developments in Reactor Physics Shielding, Kiamesha Lake, New York, September 12-15, 1972 (USAEC) p.115.

(2) T.J. Haste and M.G. Sowerby (1978) AERE-R 8961.

(3) G. Carraro and W. Colar (1970). Proc. Int. Conf. Nuclear Data for Reactors, Helsinki, IAEA-CN-26/116, 1, 403, IAEA Vienna.

(4) N.J. Bee (1981). "The relationship between the nuclear Doppler and self-shielding effects and the local s-wave strength function in ^{238}U". An invited paper presented at the Joint IAEA/NEA Consultants Meeting on Uranium and Plutonium Resonance Parameters, Vienna, September 28 - October 2, 1981.

(5) D.K. Olsen, G. de Saussure, R.B. Perez, F.C. Difilippo, R.W. Ingle and H. Weaver (1977) ORNL/TM-5915.

1.5 Background and resolution functions in neutron time-of-flight spectrometers (D.B. Syme)

Progress has been made in understanding and reducing systematic discrepancies between different measurements of neutron resonance parameters. The main difficulty has been an inadequate knowledge of the related background and resolution functions of the neutron time-of-flight spectrometers on which most measurements were made.

Background measurements have relied heavily on the well-known 'notch filter' method but this has recently been shown to give systematically erroneous results at higher energies and also when the background is of several different components (PR/NP 27, p.19) e.g. neutron capture on hydrogen nuclei in the source moderator produces a background of 2.2 MeV gamma rays which confuse measurements of background due to scattered neutrons. Due to the finite width of the strong resonances used in the notch filter method, any scattered (out-of-time) neutrons which have not suffered much change in energy are not detected as a background and their contribution must instead be calculated and included as a 'tail' on the resolution function of the spectrometer (PR/NP 27, p.20 and PR/NP 28, p.18). This effect and the multiple scattering within the detector itself lead to a final resolution function even more asymmetric than the contribution from the moderator. However it has been common to deduce resonance parameters assuming the moderator resoluton function to dominate and this has undoubtedly led to systematic errors in many cases.

The work quoted above has led to the development of a new method of treating notch filter data[(1)]. This method is valid to higher energies and determines the total background (even when it is composed of mixed radiations). It can be applied retrospectively to earlier raw data. In the same report[(1)] it has also been established that when the data have reached the stage of resonance analysis, modern shape analysis programmes can usefully confirm whether backgrounds have been properly removed from the raw data. The application of these methods should help resolve discrepancies between existing sets of resonance parameters and allow more reliable sets to be produced in future.

The following sections provide some examples of the detailed work required properly to characterise the backgrounds and resoluton function of a time-of-flight spectrometer.

1.5.1 Calculation of the resolution function for ^{6}Li glass detectors (D.B. Syme and G. Robertson (Imperial College))

The time-dependent Monte Carlo neutronics code MORSE has been used to simulate neutron detection in various arrangements of ^{6}Li glass detectors and light guides. In Fig. 1.2 is shown the resolution function component of a 2 cm polythene moderator for comparison with the results of the Monte Carlo calculations for 1 keV neutrons in a KG2L ^{6}Li glass 25 mm thick and 110 mm diameter. The inclusion of a perspex light pipe in the neutron beam

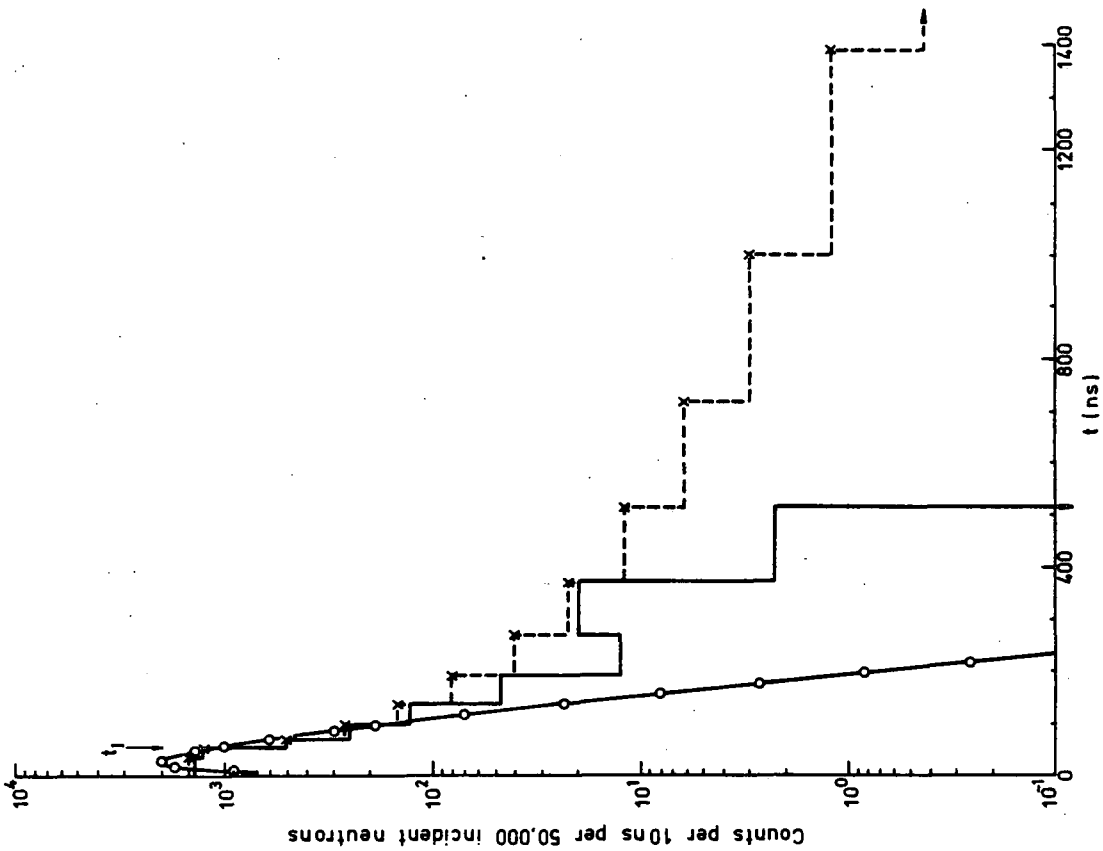

Fig. 1.2 Results of a Monte Carlo calculation of the response to 1 keV neutrons of a KG2L ^{6}Li glass detector 5 mm thick and 110 mm diameter. Full histogram - bare glass; Dashed histogram - glass with 5 mm perspex light pipe behind; Curve - typical moderator resolution function.

causes a much increased tail due to multiple scattering. For a moderator function alone, the fraction of events 'detected' after $T = 7T_m$ (= 100 ns in this case) is 1.7%. (T_m is the mean time between collisions in the moderator.) This fraction is increased to 10% for the bare ^{6}Li glass and to 18% with the 5 mm perspex light pipe. Using the wrong resolution

function in shape analysis gives neutron resonance widths systematically wrong by similar amounts.

Fig. 1.3 shows similar results for a thinner glass with 3.8 keV neutrons. The choice of a 12.5 mm thick glass with no light pipe gives a quasi-exponential tail of half life similar to that always present from the moderated source and may be considered optimum in the perpetual compromise between efficiency and resolution.

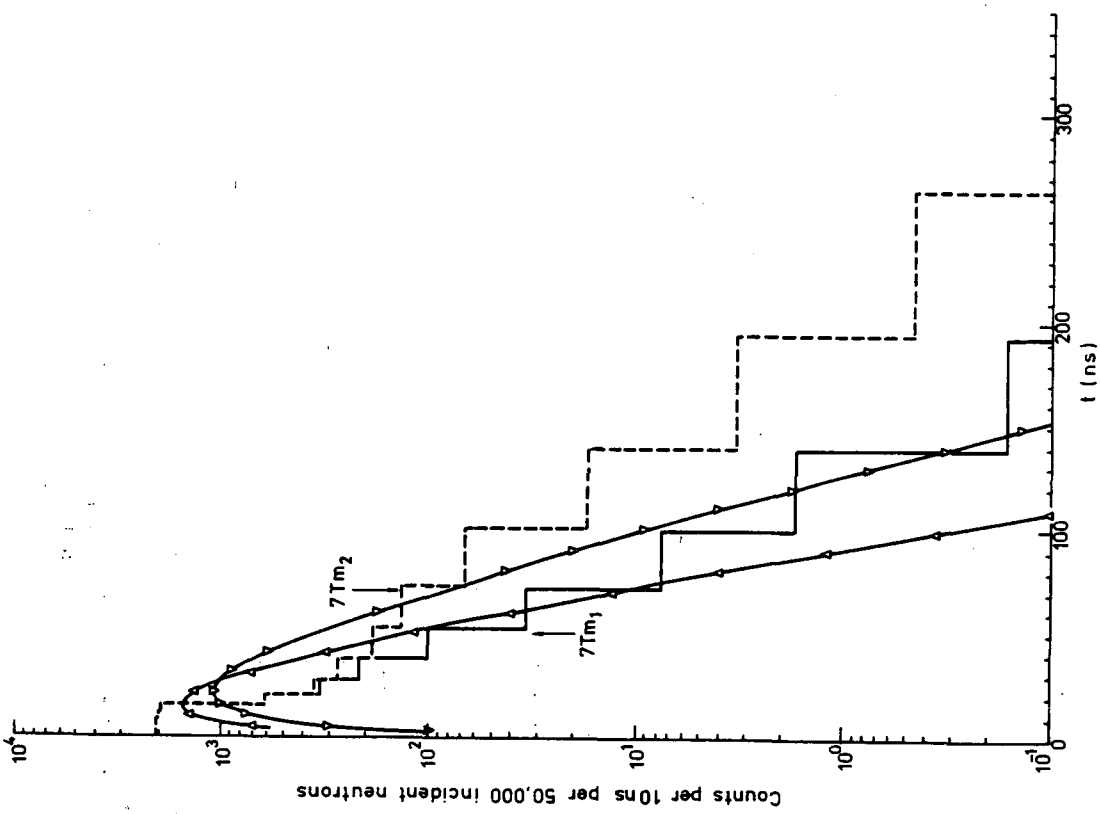

Fig. 1.3 Results of a Monte Carlo calculation of the response to 3.8 keV neutrons of a 12.5 mm thick, 110 mm diameter KG2L ^{6}Li glass detector. Full histogram - bare glass; Dashed histogram - glass with 12.5 mm quartz light pipe behind; Curves - typical moderator resolution functions for T_m = 7 and 10 ns.

1.5.2 Calculation of the effect of source room return on a spectrometer resolution function (D.B. Syme, A.D. Gadd and R.B. Thom (Imperial College))

In PR/NP 27, p.20 we described the method and some initial results for a Monte Carlo simulation of the pulsed moderated neutron source based on

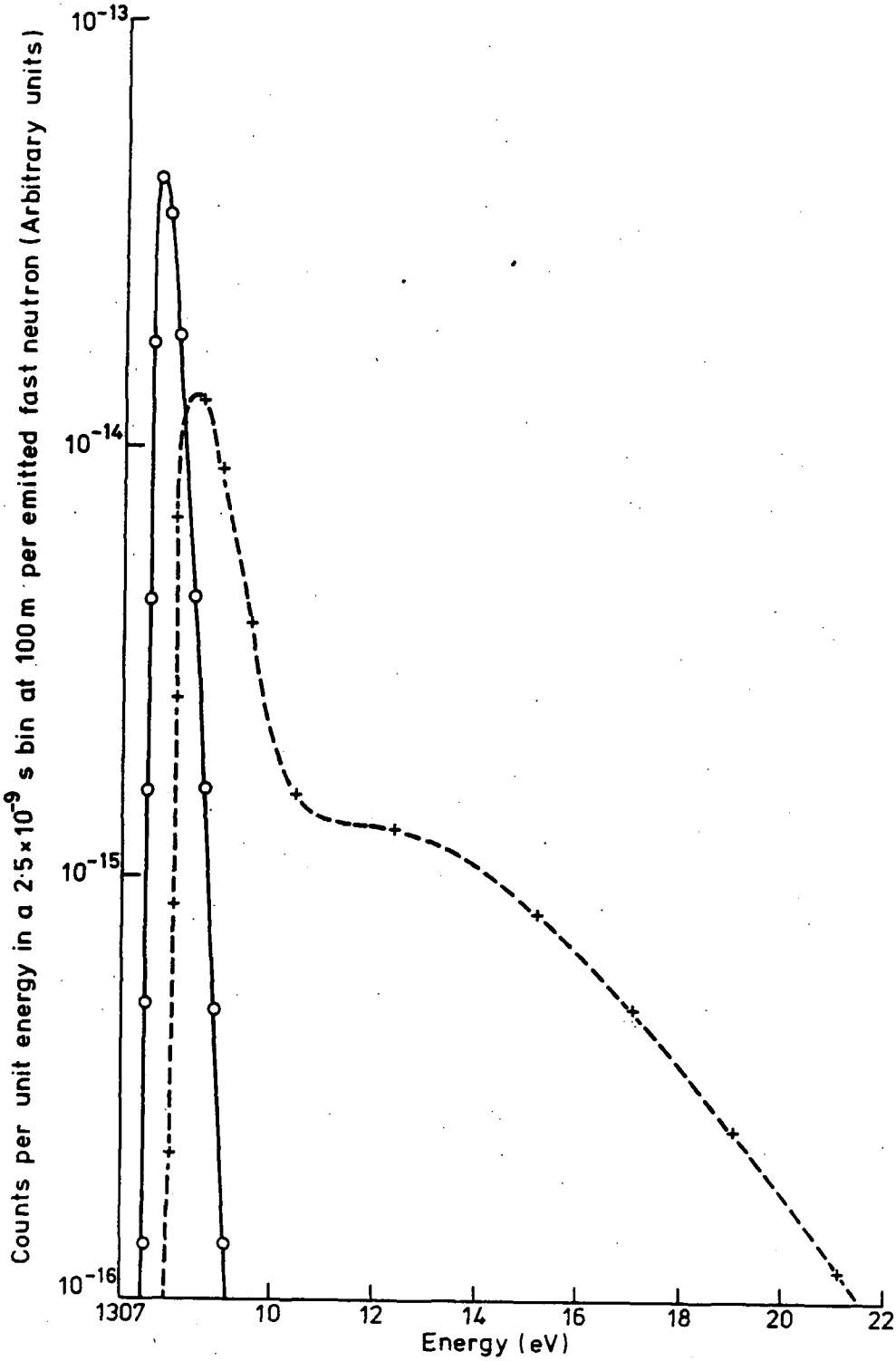

Fig. 1.4 Calculation of source room return effects for the Harwell synchrocyclotron neutron time-of-flight spectrometer. The full curve is the effective energy resolution function shape for neutrons detected in a 2.5 x 10^{-9}s wide time bin at 100 m from the moderated source, under the condition that the fast neutron burst has been moderated directly, without scattering from the source enclosure walls. The nominal energy is 1308 eV. The dashed curve is similar but for neutrons which have scattered from the source enclosure walls before moderation.

the Harwell synchrocyclotron. The calculation predicts moderated neutron spectra differential in energy and time following a burst of fast neutrons incident on the moderator directly or via a neutron scattering in the adjacent magnet poles. (The pole gap is 300 mm). In Fig. 1.4 are shown direct and pole-scattered energy spectra corresponding to a nominal neutron energy of 1308 eV.

The scattered component is nearly 10% of the prompt one and is essentially all found within a fractional energy loss of 1.6%. This tail due to 'source room return' cannot be determined by the notch filter method as this necessarily utilises resonances of width greater than 2% to remove the prompt neutrons and in this case the room-returned neutrons are themselves removed. Therefore this component must be calculated and included in the resolution function.

It has been confirmed that the intensity of the scattered tail reduces roughly as the inverse square of the source room radius and its delay and dispersion increase with the radius. Linac sources have much larger source rooms and room return should be small in these cases. It is clearly desirable to have a large detector enclosure for similar reasons. In fact conventional detectors with '1/v' response are more normally used in a heavily-shielded compact enclosure, whose effect should be included in a complete calculation of the detector response function.

1.5.3 Direct measurement of the resolution function of a neutron time-of-flight spectrometer (D.B. Syme and M.A. Wilkins)

For modern pulsed white neutron sources based on accelerators the resolution function is often dominated by the contribution from the source and its closely coupled moderator. This basic line shape is generally calculated but these calculations have seldom been confirmed by direct measurement. An arrangement has been made to enable such measurements as part of the commissioning of the fast neutron target on the new Harwell electron linear accelerator, HELIOS.

A sample of atomic weight A is placed at a flight path of 7.5 m and observed by a Moxon-Rae detector. Gamma rays are detected following neutron capture into narrow resonances of the compound nucleus $(A+1)^*$. The sample and the path length are chosen so that their contributions to the measured widths of the resonances are much less than the characteristic width of the source resolution function, which is in effect observed directly. The sensitivity of the method with various samples has been

confirmed by simulation using the known resonance parameters and the shape analysis code 'REFIT', and the apparatus is currently being assembled.

---

(1) D.B. Syme, Report AERE-R 10244 (1981).

1.6 Evaluation of neutron nuclear data

1.6.1 Nuclear Data Codes (E.M. Bowey and B.H. Patrick)

Following the implementation of codes from Brookhaven National Laboratory for ENDF/B Libraries (PR/NP 28) a second set of codes for ENDF/B processing has become available. These, written by D.E. Cullen (LLL), include a code (RECENT) to calculate pointwise cross sections from resonance parameters, SIGMA1 to Doppler broaden these cross sections, and a code EVALPLOT to provide plots of the pointwise data. A new version of SIGAR (SIGAR7) has been issued by Winfrith which performs the same tasks as RECENT plus SIGMA1, but on data in the UKNDL format. In collaboration with J.S. Story and D.E. Cullen (also M.C. Moxon using his code MLCS) a series of tests was carried out on invented resonance parameters for an imaginary non-fissionable nuclide to compare the results of calculations made in the various codes. This uncovered errors in the programs but when these were corrected the results were found to be in good agreement within the specified errors (< 0.2%). More exhaustive tests will be carried out to ensure that this agreement is maintained when the calculations are applied to fissionable nuclei.

1.6.2 Nuclear Data Files (B.H. Patrick and E.M. Bowey)

The evaluation of ^{243}Am in the resolved resonance region, referred to in PR/NP 28, p.21, was performed using an earlier version of SIGAR. It seemed appropriate therefore to rerun the resonance parameters through SIGAR7 as a check. Differences were observed which were thought to be due to errors in the Doppler broadening procedures used in the earlier version. It has been necessary therefore to construct a revised version (DFN 1010A) of the ^{243}Am evaluation. For the same reasons, a revised version of DFN 1009B (^{241}Am) has been made.

1.6.3 Calculation of Doppler broadening using the program MLCS (M.C. Moxon)

In the multi-purpose resonance program REFIT Doppler broadened cross sections are calculated using a multi-level formalism and an ideal gas model. These subroutines have been extracted and used to produce a program MLCS which gives Doppler broadened cross sections as output.

Comparisons of the cross sections given by MLCS with SIGAR and the Cullen program SIGMA1 showed up discrepancies between the different codes. This was shown to be due mainly to inadequate accuracy in the energy grid used in MLCS for calculating non-Doppler broadened cross sections. Discrepancies appeared when the width of a resonance was less than $\sim 10^{-5}$ of its energy. The use of double word length for the energy grids solved the problem and we are at present incorporating the latest version of MLCS into the analysis program REFIT.

1.6.4 Joint Nuclear Data File (M.G. Sowerby and M.C. Moxon)

The U.K. is participating in the creation of the European-Japanese Joint Nuclear Data File of neutron cross sections for reactor applications. As an initial contribution to the project, the existing evaluations of natural iron and ^{238}U have been examined and recommendations made of the evaluations to be included in the file. The present status of the measurements and evaluations of resolved and unresolved resonance parameters of ^{238}U has been presented to the IAEA Consultants Meeting on Uranium and Plutonium Resonance Parameters, Vienna, 1981. (See also section 1.4).

1.6.5 Analysis of correlated experimental data (M.G. Sowerby and M. Bailey)

In most analyses of experimental data it is assumed that the errors on data points are uncorrelated. It is obvious that in many cases this is incorrect and it has been shown[1] that the neglect of correlations can lead to erroneous results. The ENDF/B-V format for evaluated neutron cross section data includes covariance data and it is important that experimentalists provide the information that the evaluators require to generate these. This whole problem was addressed in a talk given to the Neutron Interlaboratory Seminar held in Oxford in July 1981 and a paper on the topic is being prepared. In addition a computer code has been written which applies the least squares method to correlated data which depend linearly on the parameters to be estimated[2].

1.6.6 The status of the FISPIN code on Harwell computer (D.A.J. Endacott FISPIN coding.

During the year a new version of the FISPIN code has been implemented at Harwell. As it differs from the present version 5 only in minor details, it is labelled version 5.1.

FISPIN Data Libraries. Updated versions of the cross section data used in the code for PWR, CAGR, MAGNOX, BWR and Fast Reactor systems have been installed on the computer. For the thermal reactor systems irradiation-dependent cross sections are provided.

A macro program for compiling the correct input parameters when using the above Data Libraries has been written and is at present being tested. The use of this should help to prevent incorrect input data being used with the code; this is at present a frequent source of user error.

A set of standard test cases has been run with version 5.1 together with these new Data Libraries and the results compared with similar tests run on the Risley computer. The results are essentially identical.

---

(1) F.J.G. Perey, Proceedings of Conf. on Neutron Physics and Nuclear Data for Reactors, Harwell, September 1978, p.104.

(2) B.R. Martin, "Statistics for Physicists", Academic Press, London (1971).

1.7 The $^{93}Nb(n,n')^{93m}Nb$ reaction (D.B. Gayther and C.A. Uttley, K. Randle (University of Birmingham), W.H. Taylor and M.F. Murphy (A.E.E., Winfrith)

(Relevant to request number 718)

The reaction for the production of the 30 keV isomeric state in ^{93}Nb is important as a monitor of damage fluence in power reactors because it is expected to have a cross section which varies with neutron energy in a similar way to the damage function and the metastable state decays with a half-life of about 16 y thus time-integrating the neutron flux. The long half-life combined with decay by low energy (~16 keV) X-ray emission necessitates the irradiation of pure niobium foils in high neutron fluences in order to measure the absolute activity induced in thin (~0.5 mg/cm^2) deposits of the irradiated material to an accuracy of about 5%. For this reason a trial run has been carried out at a mean neutron energy of 3.15 MeV on the 3 MV Dynamitron at the Birmingham Radiation Centre.

The niobium foil was placed 2 cm from a water cooled titanium deuteride target and backed by a 121 μg/cm^2 thick deposit of ^{235}U of the same size on the front wall of a fission chamber. The fission chamber monitored the neutron fluence through the niobium foil and was augmented in this measurement using the $^{58}Ni(n,p)^{58}Co$ reaction by including a thin

nickel foil with the niobium sample. The irradiation was carried out using ~150 μa of 1 MeV D_2^+ beam on the deuteride target, but the calculated neutron output based on the number of fission events per mC of beam on target was a factor of 5 lower than expected from the manufacturers stated target thickness. The low neutron output from the target was confirmed by the measured ^{58}Co activity, which gave a $^{58}Ni(n,p)^{58}Co$ to $^{235}U(n,f)$ cross section ratio in agreement with the value expected from the evaluated data. A measurable niobium activity has been detected despite the low neutron fluence, although a sufficiently accurate cross section will not be attainable from this test run. The prospects for future measurements are still hopeful provided the expected increase of approximately six in the intensity of the neutron beam can be achieved.

1.8 Fission chambers for the intercomparison of fast neutron flux density measurements (D.B. Gayther)

The two chambers, whose specifications were described in progress report PR/NP 26 (p.35-37), have now been assembled and tested with californium and Van de Graaff neutron sources. These tests show that the design specification has been achieved. In particular, measurements with the Van de Graaff IBIS in its pulsed mode showed that, using commercially available electronics, the chambers have a time resolution of 6 ns (FWHM). A preliminary measurement made in collaboration with Dr. J.B. Hunt on the 3.5 MV pulsed Van de Graaff at the National Physical Laboratory (NPL) revealed no problems. The intercomparison will commence in 1982 and definite participants will be: Bureau International des Poids et Mèsures (BIPM); Physikalisch-Technische Bundesanstalt, Braunschweig (PTB); Bruyères-le-Châtel; Electrotechnical Laboratory, Tokyo (ETL); National Research Council, Ottawa (NRC); the National Physical Laboratory and Harwell. It is hoped that additional laboratories in the United States and Russia will participate. At the present time a code of practice for using the instruments is being written.

1.9A (n,α) cross sections (J.A. Cookson and M.A. Langton*)
(Relevant to request numbers 367-8, 569-75)

This programme of work, which has been mentioned in previous reports, has continued. Various improvements which have been made to the counter telescope and electronic system have been tested and the system is now believed to be capable of giving reliable data.

---

*Physics Department, University of Birmingham

200 keV deuterons from the Cockcroft-Walton accelerator bombard a tritiated titanium target and emit 14.5 MeV neutrons at 45°. The neutron flux is monitored by measurement of alphas emitted from the T(d,n)α reaction.

A 26 mm diameter thin metal foil is mounted within a counter telescope about 90 mm from the source of neutrons. The energy spectrum of emitted alphas is recorded for various orientations of the target and telescope so that the angular distribution of the (n,α) reaction can be obtained. To date, measurements have been made on natural Al, V and Ni targets. Fig. 1.5 shows a preliminary angular distribution of the (n,α) cross section for the Al target. Other targets yet to be measured include natural Cr and Fe.

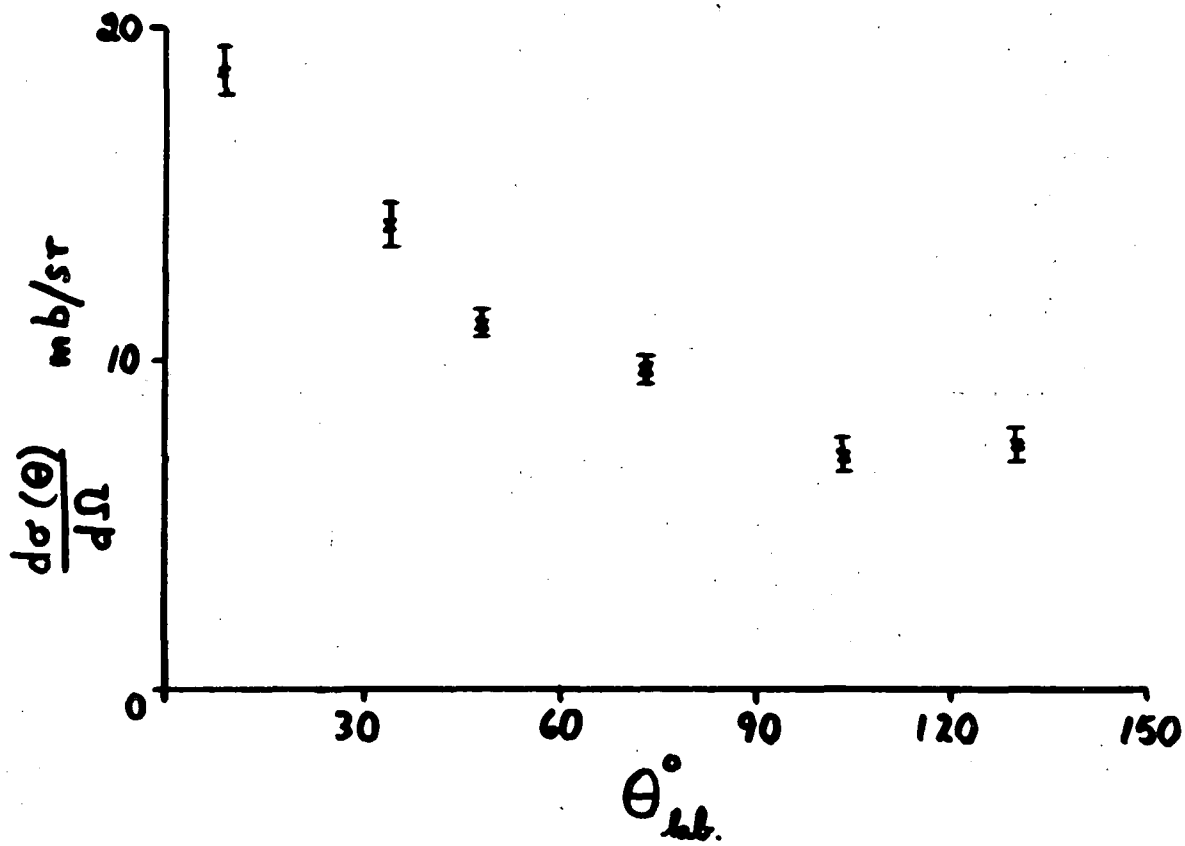

Fig. 1.5 Preliminary cross section values for the $^{27}Al(n,\alpha)$ reactions at a neutron energy of 14.5 MeV. Errors shown are purely statistical.

1.10 Selection of low-activity elements for inclusion in structural materials for fusion reactors (O.N. Jarvis)

It is desirable that the radioactivity of waste materials from fusion reactors should decay quickly to a safe level so that material recycling scenarios may be considered. A computational study has been undertaken to classify those elements which might be incorporated in a structural material in terms of the maximum allowable concentration compatible with unshielded handling after a decay period of 100 years. The acceptability of the 39 elements examined may be summarised as follows:

Primary constituents:	C, Mg, P, S, V, Cr, Y, Ta, W, Tl
Major constituents (20-50%):	O, F, Si, Mn, Fe, Pb
Minor constituents (0.1-20%):	Li, Be, B, Na, Cl, Cd, Ti, Co, Zr, In
Acceptable impurities (10-500 ppm):	Na, Al, Ar, K, Sc, Ni, Cu, Zn, Mo, Cd, Sn
Unacceptable impurities (<10 ppm):	Nb, Ag

The above results are obtained assuming that there is no chemical processing to remove undesirable impurities.

1.11 Safeguards research

1.11.1 The design of neutron die-away chambers (T.L. Morgan and B.H. Armitage

Following the construction of the neutron die-away interrogation system at Los Alamos by Kunz et al[(1)], we are undertaking a calculational study of the characteristics of neutron die-away chambers. In neutron die-away interrogation, a pulsed source of neutrons is placed inside an enclosed or semi-enclosed volume and 14 MeV neutrons from the pulse become thermalised and are repeatedly reflected by the walls of the chamber. By counting fast fission neutrons from thermal neutron interrogation, such systems can be used to assay fissile material with milligram sensitivity to ^{235}U and ^{239}Pu. The fast fission neutrons are detected by BF_3 detectors surrounded by polyethylene and wrapped in Cd; consequently, they are insensitive to thermal neutrons and detect only fast fission neutrons in the period between successive pulses of 14 MeV neutrons.

The present work is concerned with totally enclosed systems of sufficient size to accommodate 208ℓ barrels (such as are used for low level waste or scrap). The study has been confined to the use of graphite and polyethylene as structural materials. Calculations have been made of the

fission neutron rates for mixed samples of ^{235}U and ^{239}Pu within the chamber, using the computer code MONK. The code has also been used to calculate the efficiency of the detectors. The calculations thus allow the detector response to be obtained as a function of the thicknesses of the inner graphite wall, the outer polyethylene wall and the embedding depth of the fast neutron detectors.

Sufficient data have already been obtained to choose design parameters and to begin construction of a 208ℓ barrel die-away system. The chamber will have an inner 15 cm wall of graphite and an outer polyethylene wall of thickness 5 cm. Provision has been made to accommodate fast neutron detectors in four of the chamber walls and access will be via a sliding door which comprises one complete side of the chamber.

1.11.2 An examination of a package monitor for observing enriched UF_6 in undeclared packages (B.H. Armitage)

To safeguard Centrifuge Enrichment Plants it may be necessary to examine large packages entering or leaving the Cascade Hall. The aim of such a device would be to ensure that such packages do not contain uranium which may be feed or product of a clandestine enrichment plant within the legitimate Cascade Hall.

The package monitor consists of a cavity (~2 m^3), structural materials, a detector system and a pulsed neutron source. The neutron source chosen for this application is a compact neutron tube generator which has a negligible neutron background between pulses, can be placed within the cavity and produces 14 MeV neutrons with an intensity of 10^8 s^{-1}.

The neutron source is used in two modes. The first mode is delayed neutron interrogation in which the UF_6 is irradiated for a period of 7 s followed by a further 7 s period during which delayed neutrons from fission are detected. The second mode embodies some of the features of the pulsed thermal interrogation system developed by Kunz et al[1] and has been extended to detect attempts to conceal fissile material within thermal neutron shields of Cd or boron or fast neutron shields of polythene. Here a short pulse of neutrons (say 10^{-5} s) is used to detect the presence of concealment shielding within the package because as the neutron pulse gradually dies away, following thermalisation and successive reflections from the chamber walls, the presence of concealment shielding will enhance the removal of neutrons (mainly by capture) and thus reduce the die-away time.

The computer code MONK was used to calculate the number of fissions induced in the UF_6 sample when surrounded by shielding materials and contained in a rectangular wooden package. The package monitor consists basically of a polyethylene structure (Fig. 1.6) containing twenty BF_3 detectors. The MONK code was also used to calculate the number of delayed neutrons from fission counted by the BF_3 detector system. Although the calculations made with the polyethylene shielding indicate that delayed fission neutrons can be suppressed by a certain thickness of polyethythene, such a quantity of shielding material will result in a measurable change in the neutron die-away time.

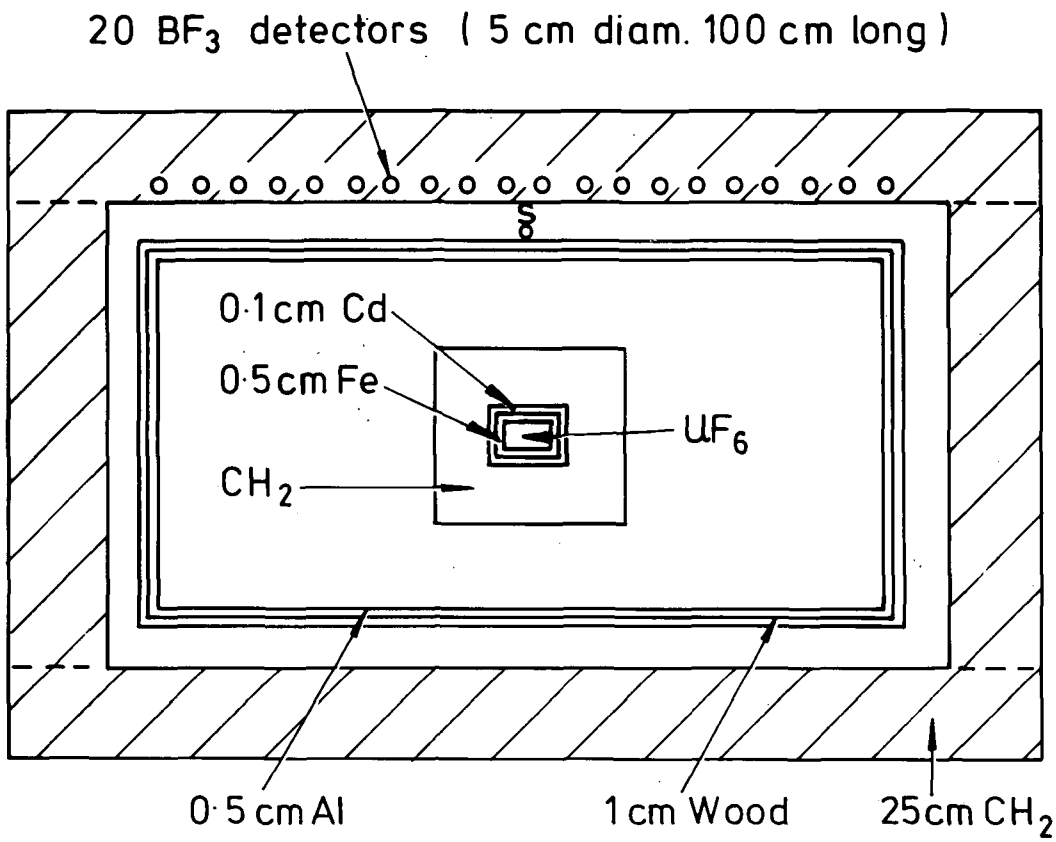

Fig. 1.6 Geometrical arrangement of UF_6 sample, neutron shielding, package and package monitor used in MONK calculations

Supporting evidence for this is provided from measurements made with a chamber of smaller dimensions (18 x 25 x 25 cm). In these experiments it was found that 0.35 kg of polyethylene produced a reduction in die-away time of about 2.5%. A conservative estimate for the equivalent amount of

polyethylene for the 2 m^3 package monitor results in a quantity which would still permit the observation of delayed neutrons from fission.

This approach to the problem of detecting attempts to conceal UF_6 has produced an encouraging result, and further work is required to validate the conclusions.

---

(1) W.E. Kunz et al, LASC report LA-UR 81-1358.

1.11.3 The determination of the ^{235}U enrichment of UF_6 gas inside pipes at the Capenhurst Centrifuge Plant (T.W. Packer)

A preliminary investigation has been carried out into the possibility of producing an instrument that will measure the ^{235}U enrichment of UF_6 gas at low pressures in pipes at the Capenhurst Centrifuge Plant.

It will be necessary to make two measurements, one to determine the mass of ^{235}U and the other to measure the total mass of uranium.

It has been shown that it is possible to determine the mass of ^{235}U by measuring the number of emitted 185 keV gamma-rays. Depending on the pressure of the gas inside the pipe, and the mass of any uranium deposited on the inside of the pipe, the total mass of uranium will be determined by using either a low energy gamma-ray transmission gauge or by exciting characteristic uranium K X-rays with 122, 136 keV gamma-rays emitted by a ^{57}Co radioisotope source. The mass of any uranium deposited on the pipe can alternatively be determined by measuring the number of 1001 keV gamma-rays emitted by ^{234m}Pa, a daughter of ^{238}U.

1.12D Neutron yields from (α,n) reactions in the light elements (D. West and A.C. Sherwood)
(Relevant to request numbers 178, 214-18, 276 and 291)

The neutron yields measured prior to the Tandem Generator shutdown in April 1980 have been evaluated and the results are now in process of preparation for publication during the course of which several amendments have become necessary.

(a) NPL announced during the year that their neutron source strength determinations should be multiplied by a factor 1.005[1] due to the presence of a previously undetected impurity in their $MnSO_4$.

(b) An exhaustive evaluation of the γ-ray spectra (from $\alpha,n\gamma$), $(\alpha,p\gamma)$ and $(\alpha,\alpha'\gamma)$ reactions), taken to detect possible neutron producing impurities in the targets, has revealed the following:

In the majority of targets no γ-rays from impurities were found and the limits set by their absence indicated that for many of the common impurities the net neutron contribution must total less than 1% of that emerging from the targets.

In the UO_2 and UC targets impurities (Si, Al, C, Mg, O) are present and together contribute about 2% to the neutron yields at 5.5 MeV. For zirconium, the entire neutron yield at alpha particle energies below 8.5 MeV was due to zxygen with some contribution from alumium and carbon and a trace of silicon. The zirconium yield measurements must therefore be withdrawn.

For stainless steel, impurities of Si, Al and Mg contribute more than 10% to the measured neutron yields at alpha particle energies below 6.5 MeV but the amounts of silicon and aluminium are compatible with the normal composition specified for the M316 alloy.

(c) A computational error has been found in the evaluation of the stopping power ratio for U and C displayed in Fig. 1.5 PR/NP 27, p.29 which was deduced from the measured neutron yields in UC and C. Data extending above 9 MeV have been incorporated and it now appears that the stopping power ratio would fit the tabulations of Ziegler[2] well if the composition of the uranium carbide were $UC_{1.12}$. An analysis of the uranium carbide target by (p,γ) has been requested.

The oxygen/beryllium stopping power ratio deduced from (α,n) yields in BeO and Be agrees with Ziegler's values to within 2% over the energy range 4 - 9.5 MeV.

The (α,n) yields from boron, oxygen in oxides, fluorine in fluorides, nitrogen in nitrides, and Li, Na and and Ca (PR/NP28, p. 34) will not now be measured since the programme on (α,n) yields has been terminated.

---

(1) E.J. Axton, private communication Oct. 1981.

(2) J.F. Ziegler, Helium: Stopping power and ranges in all elemental matter (the stopping and ranges of ions in matter. Volume 4). Pergamon Press (1977).

1.13 Comparison of the measured and calculated neutron outputs from Zebra fuel and $^{241}AmO_2$ (E.W. Lees)

The aim of this work was to measure experimentally the neutron yield to a few per cent accuracy and to compare this value with that calculated from basic nuclear data properties and from inventory codes such as FISPIN[1]. The work logically follows on from last years measurements on a mixed oxide fuel pin[2] and the experimental method is identical to that used previously. All measurements were performed using the BF_3 detector assembly designed for photofission and photoneutron studies[3].

(a) Neutron yield from unirradiated Zebra fuel

Since this work is in course of publication (AERE-R 10357), only the briefest outline will be given. The unirradiated Zebra fuel was in the form of a Pu metal plate, a mixed Pu-U oxide plate and a mixed Pu-U oxide fuel pin. Their properties were supplied by A.E.E., Winfrith and are listed in Table 1.1.

The neutron yields measured in the BF_3 detector were corrected for the effects of background and dead-time effects (<0.3%). The resultant neutron rates were then corrected for both the effect of neutron multiplication from neutrons reflected by the moderator of the BF_3 detector assembly and also for fast neutron multiplication occurring within the fuel (between 2 and 7.5%). The final statistical accuracy was ~1.5%.

The nuclear data information given in Table 4 of reference (2) was used to calculate the neutrons arising from spontaneous fission (SF). The contribution of neutrons from (α,n) reactions in ^{17}O and ^{18}O was obtained by interpolating the thick target yields in UO_2 measured as a function of α-particle energy (D. West, private communication). In all cases, (see Table 1.2), the agreement is excellent and the weighted mean of the calculated to experimental ratios is 0.997 ± 0.014 implying exact agreement to a level of 1.4% accuracy.

The range of fuel samples chosen also provides independent corroboration of the individual SF and (α,n) contributions. The Pu metal plate has ~94% of its neutrons generated from SF in ^{240}Pu, and so this measurement directly validates the revised ^{240}Pu $t_{\frac{1}{2}}$ (SF) as measured at Geel[4] (Note if the Nuclear Data Sheets value for ^{240}Pu $t_{\frac{1}{2}}$ (SF) had been used, then the calculated/experimental ratio would have been 0.890 ± 0.035 and would be in clear disagreement ($>3\sigma$)). For the Pu/U mixed oxide plate,

Table 1.1

Mass composition and physical parameters of Zebra fuel

	Mass (g)		
	Mk XIV Pu Metal Plate No. 0671	Mk IV Pu/U Oxide Plate No. IV/R/3467	Type 'D' Pu/U Oxide Pin No. ZMC 1616B
^{238}Pu	0.054	-	0.013
^{239}Pu	41.82	25.74	9.749
^{240}Pu	9.30	2.89	2.442
^{241}Pu	1.08	0.180	0.291
^{242}Pu	0.28	0.026	0.070
^{241}Am	0.405	0.208	0.133
^{234}U	0.003	0.004	0.001
^{235}U	-	0.613	0.459
^{238}U	0.012	85.62	63.06
^{237}Np	0.001	0.0006	-
O	0.006	15.47	10.24
Cu cladding	4.200	-	-
Steel cladding	9.99	17.16	12.66
Impurities	0.990*	0.085	0.042
Fuel dimensions (mm)	39.22 x39.22 x2.191	49.53 x49.53 x5.531	8.46 dia x146.2
Cu cladding thickness (mm)	0.127	-	-
Steel cladding thickness (mm)	0.381	0.373	0.381

*Only 0.024g are from light element impurities in fuel

Table 1.2

Calculated neutron production per second from Zebra fuel

Isotope	Mk XIV Pu Metal Plate No. 0671		Mk IV Pu/U Oxide Plate No.IV/R/3467		Type 'D' Pu/U Oxide Pin No. ZMC 1616B	
	SF	(α,n)	SF	(α,n)	SF	(α,n)
^{238}Pu	139	-	-	-	33	181
^{239}Pu	1	-	1	1027	-	389
^{240}Pu	9533	-	2963	427	2503	361
^{241}Pu	-	-	-	-	-	-
^{242}Pu	474	-	44	-	118	-
^{241}Am	-	-	-	575	-	368
^{234}U	-	-	-	-	-	-
^{235}U	-	-	-	-	-	-
^{238}U	-	-	1	-	1	-
^{237}Np	-	-	-	-	-	-
Sub Total	10147	0	3009	2029	2655	1299
Total (n s^{-1})	10147		5038		3954	
Uncertainty	258		76		69	
Experimental measurement	10024±161		5116±86		3949±49	
C/E	1.012±0.030		0.985±0.022		1.001±0.021	

only ~59% of neutrons arise from ^{240}Pu SF and 40% from (α,n) reactions. Thus the good agreement for this measurement validates the (α,n) yields measured by West.

If the neutron outputs had been calculated using FISPIN[1], then for the Pu metal plate since the same nuclear data sets are used as in NDS, the value would be clearly an underestimate. However, since the (α,n) data used in FISPIN are ~20% higher in yield than the experimental measurements of West, for the UO_2/PuO_2 plate there is fortuitous agreement between the FISPIN yields and experimental measurement (C/E = 0.99).

In conclusion, this work has confirmed the findings of reference (2) viz

(a) the revised ^{240}Pu $t_{\frac{1}{2}}$ (SF) as measured at Geel is correct - the NDS evaluation is 12.9%% too large;

(b) the (α,n) yields from thick targets of UO_2 as measured by West are valid data bases for estimating the (α,n) contribution from U and Pu oxide fuels;

(c) both the (α,n) and the SF Data Libraries in FISPIN need to be updated.

(d) the nuclear data information presented in reference (2) permits the neutron outputs from U and Pu unirradiated fuels to be calculated with confidence provided that light element impurities are negligible.

(b) Neutron yield from a $^{241}AmO_2$ sample

As reported in PR/NP 26, pp. 49-50, a relatively 'pure' 3 g sample of $^{241}AmO_2$ was identified by gamma-ray spectroscopy. A delay in measuring the neutron yield of the sample was encountered due to the reconstruction and calibration of the large BF_3 detector assembly on the new Harwell linac.

The average neutron energy of the AmO_2 source was measured in the BF_3 detector assembly to be 2.4 ± 0.3 MeV by the 'ring ratio' technique[3]; this energy is intermediate between that for a ^{252}Cf fission source and an Am/B source. The measured neutron yield, corrected for background (0.13%), dead-time effects (0.40%) and source absorption (0.30%) was 9582 ± 154 n s^{-1}. For the mass of ^{241}Am present at the time of the measurement (2.988 g), this implies a yield of 3207 ± 54 n s^{-1} g^{-1}; this value is larger than our previously measured yield[5] of (2.78 ± 0.40) x 10^3 n s^{-1} g^{-1} from a 12 g AmO_2 sample and is in disagreement with the yield inferred from measurements of the (α,n) yields from thick UO_2 targets (D. West, private communication) viz 2766 n s^{-1} g^{-1} (±1%).

Table 1.3 lists the light element impurities in the 3 g sample as declared by the supplier; this information was used to calculate the

Table 1.3

Light element impurities in the 3 g $^{241}AmO_2$ sample. The maximum (α,n) contributions are calculated from the thick target yields measured by West (private communication) unless stated

Element	Declared ppm weight	Calculated maximum (α,n) contribution	Experimental limits on (α,n) contribution
Li	0.3	0**	
Be	10	297	< 40
B	10	150**	< 84
F	30	42*	<205
Na	20		
Mg	<60	30	
Al	1000	278	
Si	30	1	
S	100		
Cl	100		
V	30		
Cr	100		
Mn	20		
Fe	300	0	
Ni	20		

* From Wilson et al (LASL LA-8757-PR, p.40) calculated yield

**Scaled from Liskien and Paulsens calculated yields for Be, B and Li

expected maximum neutron yield from the available data on (α,n) yields in thick targets. Also given in Table 1.3 are limits on (α,n) contributions deduced from the measured gamma-ray spectrum[5]. Combining the information results in a maximum neutron yield of 475 n s^{-1} from light element impurities and only accounts for ~1/3 of the discrepancy.

A further search of the gamma-ray spectra at higher energies revealed clear evidence of an Al impurity (see Figs. 1.7 and 1.8). Unfortunately, the gamma-ray yields measured by West and Sherwood[6] cannot be used to identify the strength of this impurity since their measurements with an α-particle beam are sensitive to angular distribution effects. On further prompting of the supplier, it was conceded that a range of 300-3000 ppm Al was possible. At the upper end of this range, the measured neutron yield from oxide per gram of ^{241}Am after correction for light element impurities would only be different from the yield inferred from West's measurements by 95 ± 59; this is no longer inconsistent.

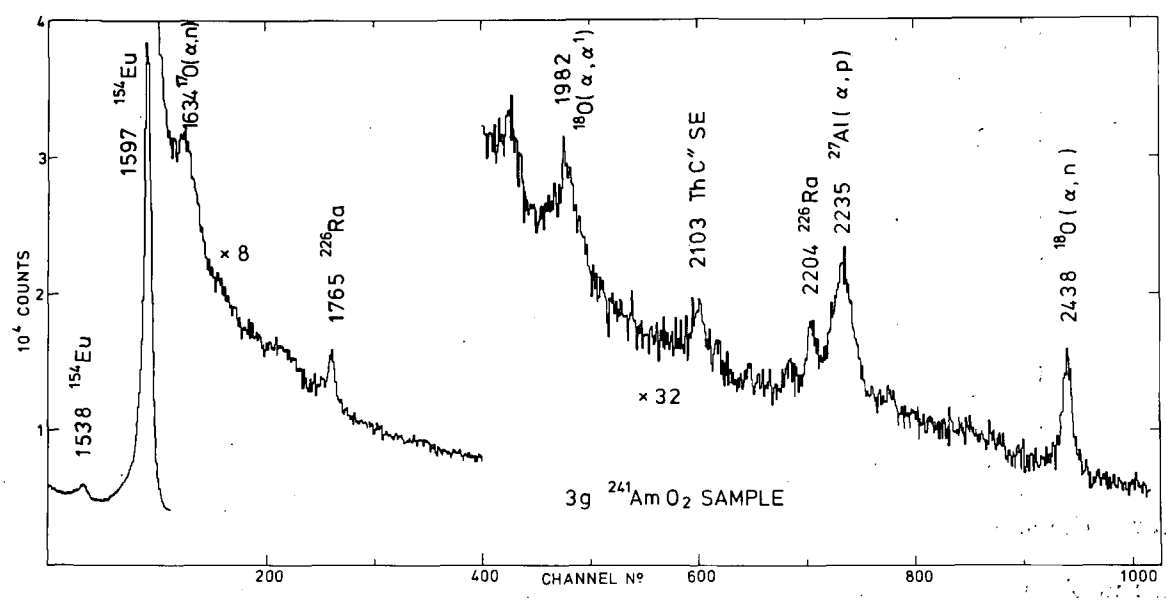

Fig. 1.7 The gamma-ray spectrum from the 3 g $^{241}AmO_2$ sample between 1.5 and 2.5 MeV. The ^{154}Eu impurity will not contribute to the (α,n) yield.

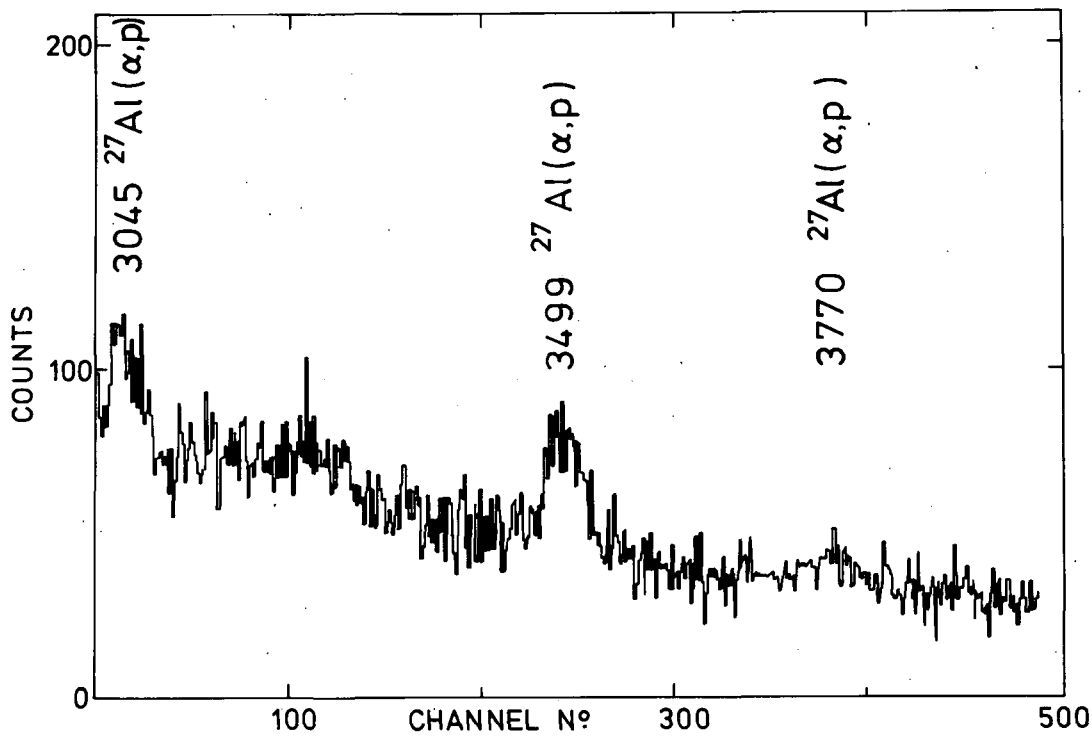

Fig. 1.8 The gamma-ray spectrum from the 3 g $^{241}AmO_2$ sample between 3 and 4 MeV.

The relevance of such measurements has now been superceded by the measurements of West, as confirmed in section 1.13 (a); no further work is envisaged with AmO_2 samples.

---

(1) R.F. Burstall, UKAEA No. R 328(R) 1979.

(2) E.W. Lees and D. West, AERE-R 10184 (1981) and PR/NP 28, pp. 35-37.

(3) E.W. Lees, B.H. Patrick and E.M. Bowey, Nucl. Instr. Meth. 171 (1980) 29.

(4) Budtz Jorgen et al, BNL 50991 (1979) 239.

(5) E.W. Lees and D. Lindley, Ann. of Nucl. Energy 5 (1978) 133.

(6) D. West and A.C. Sherwood, to be published.

1.14 Plutonium dating (D. West and A.C. Sherwood)

In the course of examining the neutron output from a fast reactor fuel pin[1] the emitted γ-rays were also examined. The interpretation of the spectrum in the hundreds of keV region has led to a possible method of dating the last chemical separation process which the plutonium underwent[2,3]. The age of a plutonium sample may prove to be of use in the future as it constitutes an additional label or "finger-print" attached to any sample of plutonium, which may be used for identification in accountancy or safeguards applications for instance.

The method depends on the presence of ^{241}Pu (half-life 14.4 years) in the mixture of plutonium isotopes. This decays by two routes, predominantly by a very soft β-ray (20.8 keV) to ^{241}Am (half-life 432 years) and in 0.00246% of cases by α-decay to ^{237}U (half-life 6.75 days). Both ^{241}Am and ^{237}U then decay (by α-emission and β-emission respectively) to ^{237}Np (half-life 2.15 x 10^6 years). The new method depends on measuring what is effectively the activity ratio of ^{237}U and ^{241}Am which is a sensitive single-valued function of the time elapsed since ^{241}Pu was freed from pre-existing ^{241}Am and ^{237}U as an incidental accompaniment to the removal of fission products and uranium during chemical processing of spent fuel.

Of course a similar objective can be achieved if the growth of ^{241}Am in a sample is measured as a function of time and the amount of ^{241}Am is then extrapolated backwards in time. However this involves measurements over a period of time comparable with the age of the sample if reasonable accuracy is to be achieved and is not practical except for very young samples. The new method of dating involves a measurement at a single time and should enable ages even up to 100 years to be assigned. Moreover, as the method makes use of γ-rays only, it is non-invasive.

The technique has so far been applied to fast reactor fuel pins (5 mm diameter) which requires the use of a sample of known age and similar configuration to avoid corrections due to self-absorption of the γ-rays. Two fuel pins M120 and M119, each containing annular $(Pu,U)O_2$ pellets, were compared. The records state that the isotopic composition of M120 was measured on 15 March 1978. No record is available of its date of separation but it was usual for an interval of 180-200 days to elapse between the two dates. The approximate date of separation for M120 was therefore 15 September 1977. M119 was stated to contain material separated

in March 1978. Both fuel pins contained 5 g of plutonium of which 3.7% was ^{241}Pu. The ages determined for fuel pin M119 on 9 April 1981 are contained in Table 1.4 using various pairs of γ-rays.

Table 1.4

Determinations of the age of fuel pin M119 on 9 April 1981 using various pairs of γ-rays and a separation date for fuel pin M120 of 15 September 1977

	208.0 keV	267.5 keV	332.4 keV
662.4 keV	3.547 ± .023 years	3.550 ± .024 years	3.549 ± .024 years
712.9 keV	3.549 ± .032 years	3.552 ± .033 years	3.551 ± .033 years

The mean age of fuel pin M119 comes out as 3.549 ± 0.019 years corresponding to a separation date of 20 September 1977 ± 7 days. Its age determined by this method is therefore identical with that of fuel pin M120. As mentioned above the records are not sufficiently precise for this to constitute a test of the technique but the error of ± 7 days on a 3.5 years old sample is representative of the precision which can be expected of the technique. Fuel pins of age about 9 years were also available but, as their pellets differed from M119 and M120 in diameter and in having no central hole, an accurate intercomparison of age was not possible. Qualitative agreement with the stated ages was obtained however.

The measurements in Table 1.4 occupied a period of some 15 hours for each pin. They were carried out with the fuel pin under 2 mm of Sn absorber to eliminate the strong 59.5 keV γ-radiation. The fuel pin was measured at a distance of 500 mm from a Ge(Li) detector of volume 71 ml. A limitation with fuel pins is the spread in age within a batch of material used in making single pins. This may amount to as much as 3 months but it should not invalidate the concept of an average age for ages appreciably greater than this and it certainly does not prevent materials from the same batch being positively identified. Efforts are in hand to obtain freshly prepared fuel pins of known age to act as standards. Thin laboratory samples of different known ages are also being prepared and will be kept to enable future comparisons to be made should interest in the technique be forthcoming.

The details of the new method of age determination can be found in the publications[2,3] already referred to. However the following special features of ^{241}Pu decay which make it eminently suitable for dating purposes should be mentioned here.

(i) The "weak" branch which decays via ^{237}U carries the predominant γ-ray yield. This makes its presence detectable in comparison with the "strong" branch (whose intensity increases with time) to much longer times than would otherwise be the case.

(ii) The lifetimes of ^{241}Pu and its decay products are such as conveniently to cover a period up to 100 years or so.

(iii) The ^{241}Pu lifetime (about which there is some uncertainty) is not involved in the activity ratio with 100% sensitivity for ages shorter than 50 years. The lifetime of ^{241}Am is, however, involved with 100% sensitivity.

(iv) The Table of Isotopes[4] now lists 21 examples of branched α, β decay similar to that occurring in ^{241}Pu. None of the other branched-decay schemes appears to have any practical importance for dating similar to that which now seems possible with ^{241}Pu.

---

(1) E.W. Lees and D. West. Report AERE-R 10184.

(2) D. West and A.C. Sherwood. Report AERE-R 10020.

(3) D. West and A.C. Sherwood. Ann. of Nucl. Energy 8 (1981) 441.

(4) C.M. Lederer and V.S. Shirley. Table of Isotopes 7th Edition (1978) John Wiley and Sons Inc.

2. CHEMICAL NUCLEAR DATA

2.1 Introduction

The main committee (Chairman: J.G. Cuninghame, AERE) has held two meetings during the year and the Decay Library Sub-committee (Chairman: B.S.J. Davies, Berkeley Nuclear Laboratories), three. The generation, updating and maintenance of the Data Library remains the most important task of the Committee. The current position is that both the fission product decay data file (UKFPDD-2) and the heavy element decay data file (UKHEDD-1) are well up to date and are probably superior to anything available elsewhere. The activation products decay data file (UKPADD-1) is now under extensive revision and expansion from the present 91 nuclides to a proposed 410 nuclides. The fission yields file will also be updated in the near future. All the above files exist in ENDF/B format on the computers at Harwell, Winfrith and Berkeley.

Members of the Committee continue to be active in measurement of heavy element and fission product decay data and of parameters such as α, but the available effort now stands at around 25% of what it was 10 years ago.

2.2 Fission Measurements

2.2.1 BIZET experiments to study the effect of change of reactor neutron spectrum on fission yields (J.G. Cuninghame and H.H. Willis (AERE))

25 fission yields have been measured for fission of ^{235}U, ^{238}U and ^{239}Pu in both inner and outer core positions in each of the two Zebra/Bizet cores BZB/3 (a conventional core arrangement) and BZD/1 (an unconventional one).

Data reduction and estimation of errors is now nearly complete. As an example, Figs. 2.1 and 2.2 show the final fission yield curves for ^{235}U fission in the inner and outer core positions of core BZB/3. It is significant that although the neutron spectrum is much "softer" in the outer position, there is no significant difference between the two curves within the ~5% errors shown.

2.2.2 Fission yield measurements at Dounreay (W. Davies and V.M. Sinclair (DNPDE)

Irradiation of the capsules of the PFR fission yield experiment (outlined on pages 43-44 of UKNDC(75)P71), should be completed in 1982.

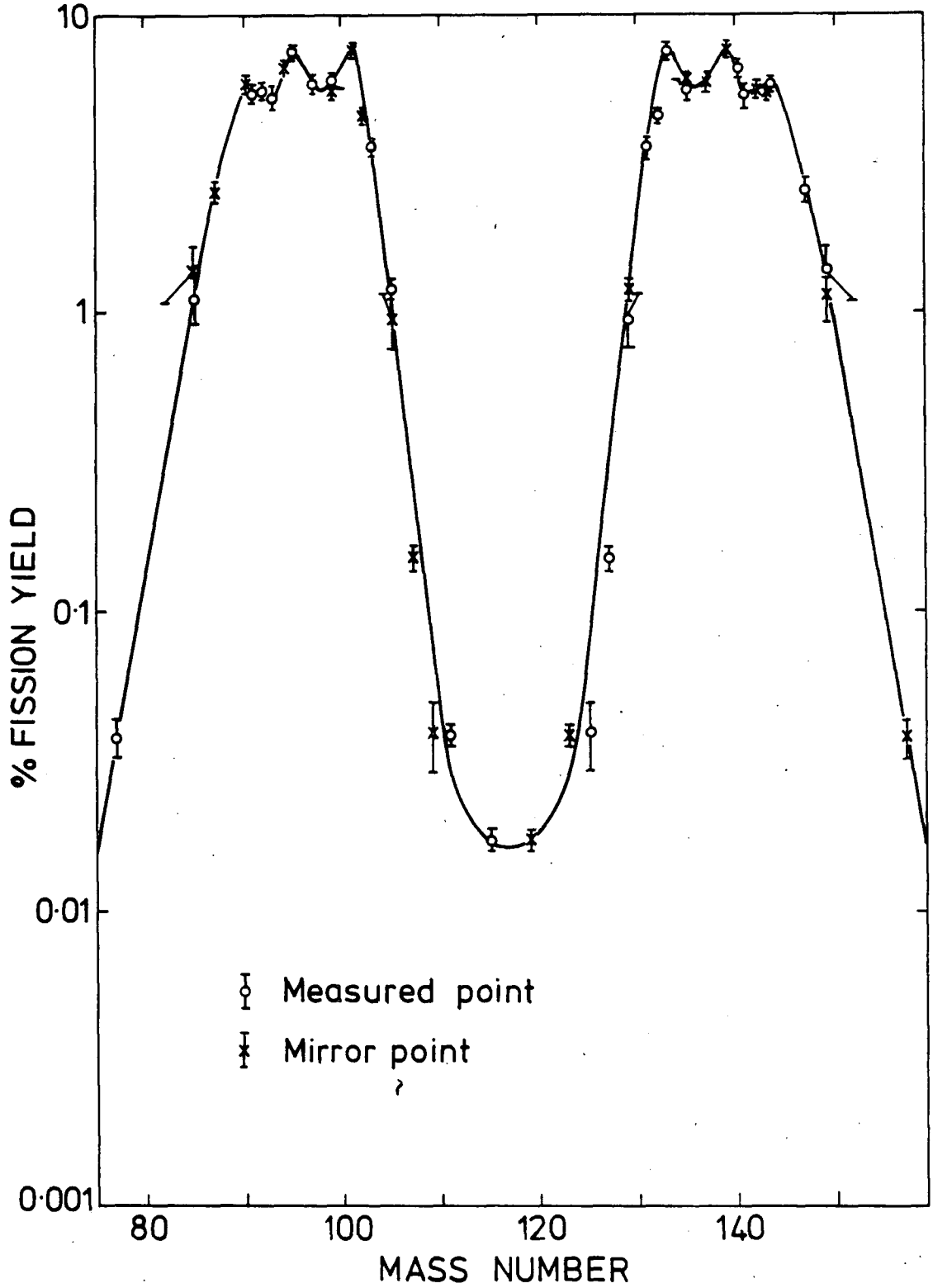

Fig. 2.1 ^{235}U fission yields; conventional core, inner core position

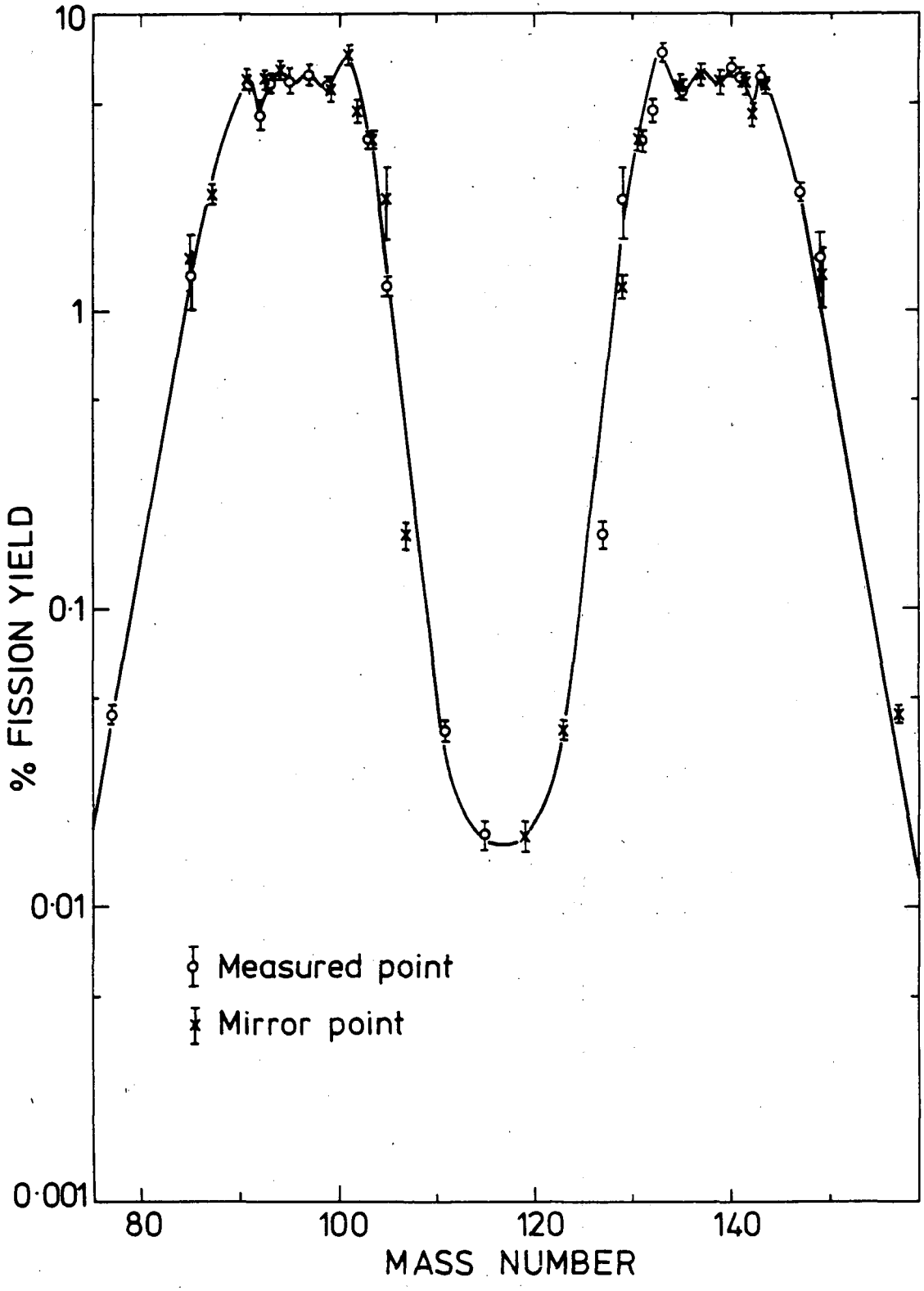

Fig. 2.2 ^{235}U fission yields; conventional core, outer core position

2.2.3 Development of the helium jet recoil transport system for VEC studies of short-lived nuclides (R. Bett, J.G. Cuninghame, H.E. Sims and H.H. Willis (AERE))

Attempts to achieve selective transport of sulphide-forming fission products by using diethyl sulphide clustering agent have not been successful. Fission products are indeed transported with good efficiency in a time of ~1/5 s, but there is no selectivity. Consequently we have now decided to concentrate on very fast chemical separations at the receiving end of the system. A generator has been built which allows us to substitute KCl for diethyl sulphide as the clustering agent; this is known to produce very high transport efficiencies. The gas handling system has been redesigned so that transport can be achieved by running the recoil chamber at up to 4 atmospheres (instead of 1 atmosphere) and the receiver at 1 atmosphere (instead of under vacuum).

We have also just brought into use a pressure recoil chamber with a built-in strong (~2 x 10^6 fissions/min) ^{252}Cf source. This source provides an alternative supply of fission products, independent of the VEC runs. It has been tested using the old gas handling system and diethyl sulphide as the clustering agent and appears to work well.

2.2.4 Heavy ion-heavy element reactions (K.M. Glover and K.J. Howard (AERE))

Testing of the on-line helium jet equipment to study heavy ion-heavy element reactions has progressed slowly due to lack of effort.

2.2.5 Software for pulse height analysis and gamma spectral analysis on a PDP 11 computer (J.G. Cuninghame, H.E. Sims (AERE); G. Hayton, E. Adlington (F. International))

γ-ray or X-ray pulse height spectra normally have to be analysed by some form of peak location and fitting program which produce peak areas for individual X- or γ-rays. Sets of peak areas from a series of spectra taken from the same radioactive source counted at different times then have to be analysed by linear or non-linear regression analysis to find the half-life and activity at some reference time.

We use the peak fitting program GAMANAL which runs on the IBM computer. A PDP11 program (DECIBM) has now been written which allows us to send raw spectra from the PDP11 computer to a HUW line file via HANS, run GAMANAL, with the data and then receive the results back to the PDP11. A second program does the same thing but via a direct line to the IBM

computer; this method involves using the RSX operating system on the PDP11, and TSO on the IBM.

A further new set of PDP11 programs (GAMRES, GAMFAT, FATAL, PLTFAT) then allows us to take GAMANAL results, build up input files and then run the linear or non-linear regression analyses on them to give us our final results.

2.2.6 A PDP11 computer program and library for identifying X- and γ-ray peaks (J.G. Cuninghame, H.E. Sims (AERE); G. Hayton (F. International))

A program (PHALIB) has been written for the PDP11/34 computer which is designed to take the drudgery out of identifying unknown X- and γ-rays found in spectroscopic measurements. The data base for the program is a reformatted version of the Darmstadt γ-ray catalogue (~40,000 γ-rays) to which we have added the K, L and M X-rays for all elements up to lawrencium. Each entry consists of the energy, isotope identification, half-life and intensity of the γ-ray (energy, type and element name for X-rays). This data base is in the form of a single file (ISOTOP.LIB), consisting of ~1100 blocks residing on an RL01 disk.

There are three modes of operation:

(a) Adding or deleting entries from the data base.

(b) Listing either:
  (i) all entries for a nuclide (or element for X-rays) or
  (ii) all entries found to lie within "windows" selected by the user. These windows are limited in mass number and half-life.

(c) Searching for entries corresponding to a list of X- or γ-ray energies either:
  (i) entered from the keyboard, or
  (ii) taken from files of results automatically produced by GAMANAL or the pulse height analyser programs.

The search is conducted with windows selected by the user. In addition to mass number and half-life, energy limits on either side of the listed energies are used.

2.2.7 Fission of ^{224}Th* formed in the reaction ^{15}N + ^{209}Bi (J.G. Cuninghame, H.E. Sims (AERE); I.S. Grant, J.D. Hemingway, P. Misaelides, G.W.A. Newton and V.J. Robinson (Manchester University))

The compound nucleus ^{224}Th has been formed in the reaction ^{15}N + ^{209}Bi at several bombarding energies between 76 and 131 MeV. Fission fragment activities have been measured for chemically separated Y, Tc, Rh, Ag and Sb; rhodium is the symmetric fragment and the other elements make up

complementary pairs. Table 2.1 give relative yields for yttrium and technetium isotopes at two bombarding energies.

Table 2.1

	119 MeV	97 Mev
Y isotope		
90 m	11	2.3
91	30	17
92	66	47
93	100	100
94	58	74
Tc isotope		
99m	1.7	0.3
101	100	100
104	53	58
105	41	

Data have been collected for all five elements at three bombarding energies, which when analysed will lead to accurate estimates of the total number of neutrons emitted in association with the thorium fission. This information is required in order to derive accurate Total Kinetic Energy (TKE) values for the primary fission fragments (using the method applied to ^{208}Po in reference 1) from counter measurements on coincident fragments. Fig. 2.3 shows preliminary mass and TKE distributions from our counter measurements, using neutron numbers estimated from an evaporation code. Unlike the case of ^{208}Po, the data show that the fragment TKE value is not proportional to Z_1Z_2 and that the width of the mass distribution is greater than expected from simple statistical considerations.

---

1. J.G. Cuninghame et al "Physics and Chemistry of Fission 1979" Vol. 1, p. 551, IAEA (1980).

2.2.8 Studies of the reaction $^{238}U(\alpha,p)^{241}Np$ (J.G. Cuninghame, H.E. Sims, H.H. Willis, B.W. Hooton (AERE), S. Holloway (Imperial College))

In the previous Progress Report we reported preliminary on-line experiments with this reaction and measurements of off-line γ-spectra from unprocessed catcher foils. Because of problems with these experiments we decided to try to establish the best irradiation parameters by chemical separation of Np from irradiated ^{238}U targets, followed by X-ray and γ-ray spectrometry.

A chemical separation procedure based on anion exchange with HCl, produces Np free of ^{238}U target material and fission products in ~10 min. from the end of bombardment. We have used this procedure after irradiations with a range of α-energies from 28 to 40 MeV and have detected a peak at 175 keV with a half-life of ~17 min which may be due to ^{241}Np. Further

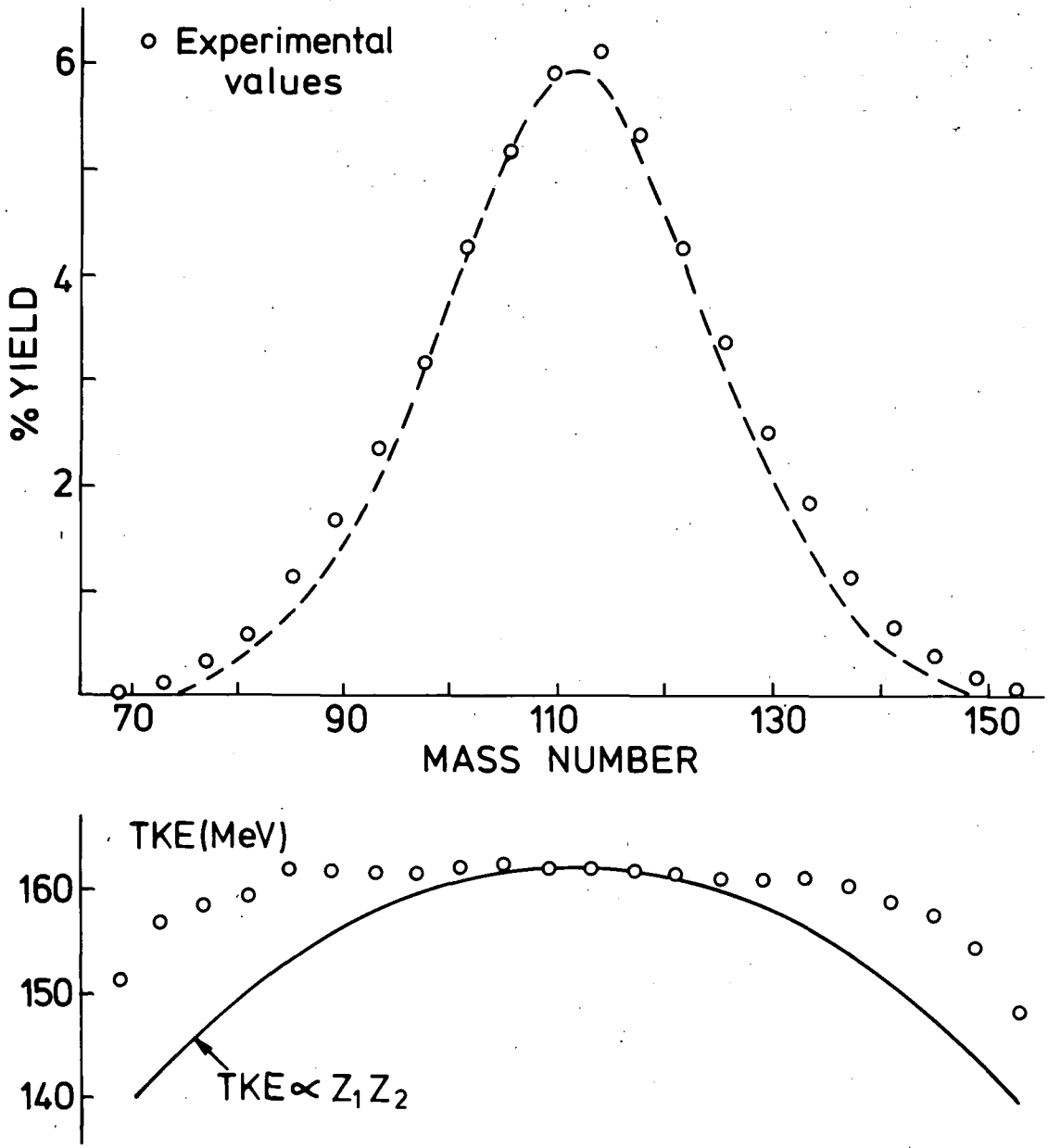

Fig. 2.3 Fission yield for ^{15}N + ^{209}Bi reaction at 76.6 MeV excitation energy. The dashed curve is the calculated primary mass distribution for excitation 40 MeV higher than the saddle point.

runs with improved chemical separation and counting methods are now under way. If these confirm that we have detected ^{241}Np, further on-line measurements will be made under the successful irradiation conditions.

2.2.9 Reactions between ^{93}Nb and ^{12}C, ^{14}N, ^{16}O and ^{20}Ne (H.E. Sims (AERE); G.W.A. Newton, V.J. Robinson, E.M. Shaw and R.J. Smith (Manchester University)

In this work yields of as many products as can be determined by off-line γ-spectroscopy (half-lives > 2 min) are measured in order to study the complex transfer (or deep inelastic) reactions which occur when nuclei are bombarded with high energy heavy ions.

Our results lead us to the following general conclusions:

(a) transfer of mass and charge from projectile to target nucleus is much more probable than the reverse. The probability of a particular mass and charge transfer P(ΔN, ΔZ) increases with ΔN and ΔZ (see Fig. 2.4). The most probable yields are consistent with rapid charge equilibration during the short interaction time, followed by a small amount of subsequent neutron evaporation from the target-like fragment.

(b) recoil momenta of target-like products are consistent with relatively large momentum loss by the projectile ("soft" collisions), which increases the complexity of the transfer reaction.

(c) excitation functions for transfer products show monotonic increases in cross section with increasing beam energy (see e.g. Fig. 2.5).

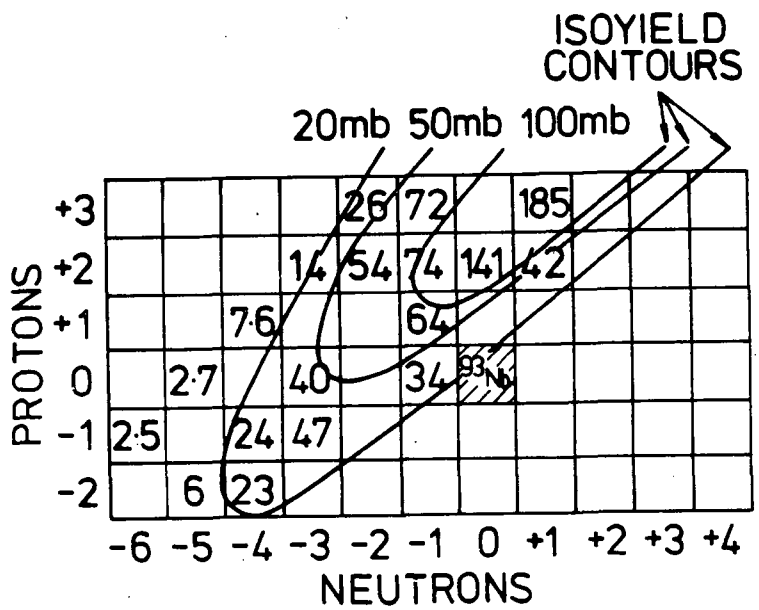

Fig.2.4 Product yields in the deep inelastic transfer from the reaction ^{16}O + ^{93}Nb with 84 MeV O ions. The numbers in the squares correspond to the relative yields for the particular products

(b) recoil momenta of target-like products are consistent with relatively large momentum loss by the projectile ("soft" collisions), which increases the complexity of the transfer reaction.

(c) excitation functions for transfer products show monotonic increases in cross section with increasing beam energy (see e.g. Fig. 2.5).

Attempts to explain these results using time-dependent Hartree-Fock calculations are currently being made by J.M. Irvine at Manchester.

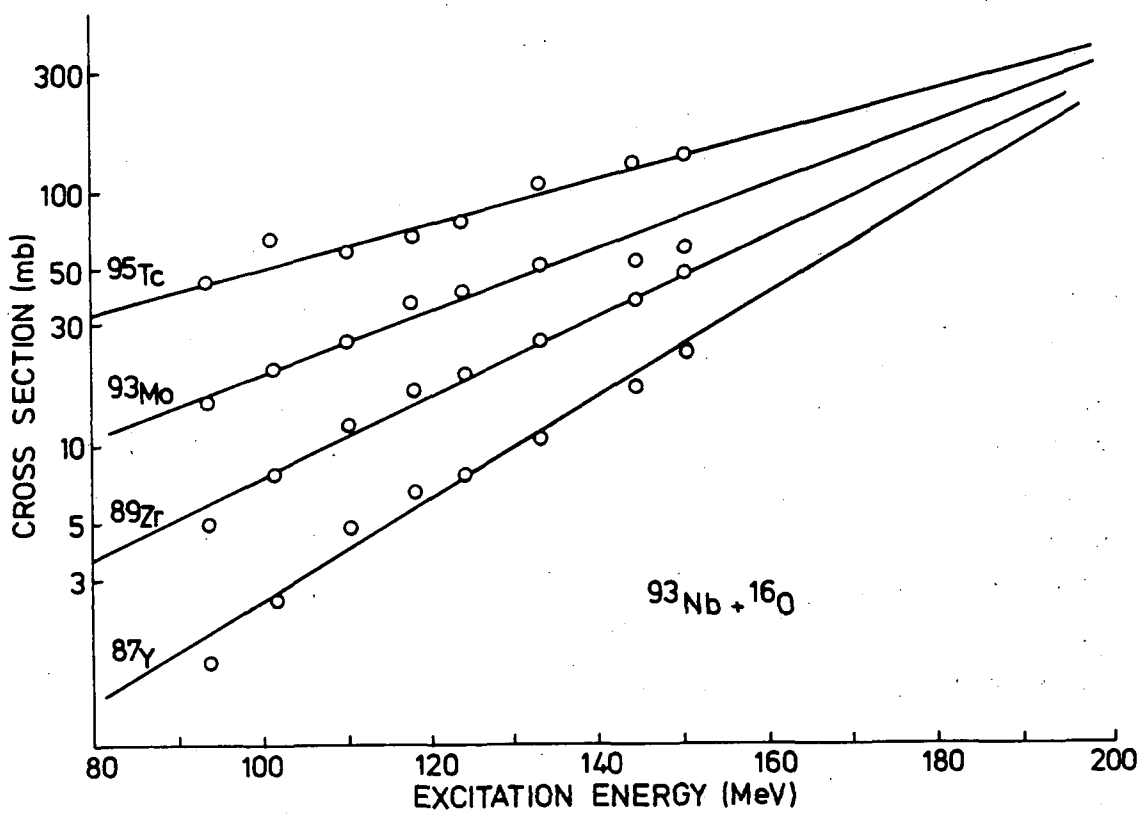

Fig. 2.5 Yields of primary fragments as a function of ^{16}O energy for the reaction $^{93}Nb + ^{16}O$

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

This work has been held up owing to reallocation of laboratory space, but has recently been restarted.

2.3 Cross Section Measurements

2.3.1 Production cross section measurements of ^{242}Cm and ^{244}Cm in core 21 in ZEBRA (D. Anstey, K.M. Glover, I.C. McKean, M. King, B. Whittaker and R.A.P. Wiltshire (AERE))

(Relevant to request numbers 1529, 1536 and 1574)

Eleven ampoules containing ^{241}Am, and four containing ^{243}Am were subjected to an extended irradiation (10.3.80 to 22.4.80) in typical fast reactor core and breeder region spectra in the ZEBRA reactor at Winfrith, to permit measurement of the cross sections leading to the production of ^{242m}Am and ^{242}Cm (from ^{241}Am), and ^{244}Cm (from ^{243}Am). The resulting ^{242}Cm and ^{244}Cm were separated from ^{241}Am and ^{243}Am respectively, by radiochemical techniques using ion exchange[1]. The small number of ^{243}Am samples available for the irradiation meant that only one sample could be irradiated in each core position in a block with two ^{241}Am samples. To ensure a duplicate set of results for each irradiation position, after dissolution the ^{243}Am samples were each divided into two fractions which were then treated independently. One of the ^{241}Am samples was similarly treated. Interim results are shown in Table 2.2 for ^{242}Cm and ^{244}Cm production. Measurements of the ^{242m}Am production cross section will of necessity be made over an extended period. Samples 6 and 12, which each contained approximately 50 mg ^{241}Am, give significantly lower values for the ^{242}Cm production cross section than the other ^{241}Am samples (ca. 10 mg) irradiated in the same block. In the ^{244}Cm production cross section measurements, corrections were applied for ^{242}Cm in-growth from the ^{242m}Am present in the ^{243}Am prior to irradiation.

---

(1) R.A.P. Wiltshire, K.M. Glover. Radiochemical techniques used to measure the cross sections leading to the production of ^{242}Cm and ^{244}Cm in ^{241}Am and ^{243}Am samples irradiated in the ZEBRA reactor at Winfrith. J. Radioanal. Chem. 64, 47 (1981) (U).

2.3.2 Measurement of neutron cross section in PFR (E.A.C. Crouch, J.G. Cuninghame, I.C. McKean and H.H. Willis (AERE), W. Davies, D.J. Lord and V.M. Sinclair (DNPDE), N.R. Daly (AWRE))

Details of this programme which will check cross section data in nuclear spectra similar to CFR, were given in UKNDC(75)P71, page 53. The irradiated capsules, which contain ^{10}B, ^{235}U, ^{238}U, ^{240}Pu, ^{241}Pu, ^{242}Pu, ^{241}Am and ^{243}Am have been dispatched to the Analytical Laboratories at DNPDE, AWRE and AERE. The Dounreay analytical work is almost complete.

Table 2.2

^{242}Cm and ^{244}Cm production cross section interim data ZEBRA core 21

Nuclide	Ampoule Number	ZEBRA Core Position	Production Cross Section (barns)
^{241}Am	1 2	50, 38) 50, 38)	2.195 ± 0.1%
^{243}Am	21A 21B	50, 38 54, 38	2.79 2.78
^{241}Am	3 4	54, 34) 54, 34)	1.149 ± 0.2%
^{243}Am	22A 22B	54, 34 54, 34	1.36 1.35
^{241}Am	7 8	50, 45) 50, 45)	4.065 ± 0.5%
^{243}Am	20A 20B	50, 45 50, 45	4.78 4.91
^{241}Am	18A 18B 6	46, 34) 46, 34) 46, 34	1.162 ± 0.5% 1.117
^{243}Am	23A 23B	46, 34 46, 34	1.27 1.30
^{241}Am	16 17 12	52, 44) 52, 44) 52, 44	3.927 ± 1.0% 3.674

2.3.3 PFR high power reaction rate experiment (K.M. Glover, R.A.P. Wiltshire and B. Whittaker (AERE))

The ^{241}Am and ^{243}Am samples which were irradiated in a high power run in PFR are now at Harwell and have been decanned and identified. It is anticipated that processing will commence early in 1982. These samples should provide complementary integral cross section data to those obtained from the ZEBRA irradiations.

2.4 Half-life Measurements

2.4.1 ^{237}Np half-life and I_α (D. Brown, K.M. Glover and B. Whittaker (AERE))

(Relevant to request number 1287)

The suitability of the stoichiometric compound $NaNpO_2(CH_3COO)_3$ for half-life measurements has been evaluated by preparing deposits ranging from 10 to 100 microgrammes per sq. in. on stainless steel substrates. The

resolution of the deposits was measured by alpha spectrometry. A similar set of sources was prepared on tantalum substrates from $Cs_2NpO_2Cl_4$. In all cases the $NaNpO_2(CH_3COO)_3$ sources were of much better quality than those prepared from the caesium compound. It is planned to start ^{237}Np half-life measurements early in 1982.

2.4.2 ^{235}Np decay data (B. Whittaker (AERE))

The half-life of ^{235}Np will be measured by gamma spectroscopy on solution samples spiked with ^{241}Am as an internal standard. ^{235}Np decays by electron capture (>99%) to ^{235}U, generating characteristic uranium K and L X-rays. The 59.5 keV ^{241}Am peak is conveniently situated in the centre of the uranium X-ray region. Decay measurements will be made by comparing the ratio of the $UK_{\alpha 1} + K_{\alpha 2}$ peaks with the 59.5 keV ^{241}Am peak over 1-2 years.

2.4.3 ^{239}Pu half-life evaluation (K.M. Glover (AERE) and A.L. Nicholls (AEE))

A comprehensive evaluation of all the ^{239}Pu half-life measuremnets published in the open literature was submitted to CNDC and to DIDWG for discussion[1].

---

(1) K.M. Glover, A.L. Nicholls. A critical review and evaluation of published ^{239}Pu half-life data. DIDWG(80)/P229 CNDC(80)P5.

2.5 Fission Product Decay Data

2.5.1 Data Library Sub-committee

Data Library Sub-committee members: B.S.J. Davies (Chairman, CEGB), A. Tobias (Secretary, CEGB), B. Aldred (BNFL), K.M. Glover (AERE), M.F. James (AEEW), A.L. Nicholls (AEEW), H.E. Sims (AERE), D.G. Vallis (AWRE).

The Sub-committee is responsible for the development and maintenance of the data files which together constitute the UK Chemical Nuclear Data Library, the current status of which is shown in Table. 2.3.

A draft report (AEEW-R 1407) has been prepared describing and listing the contents of the heavy element decay data file (UKHEDD-1). The decay data are in ENDF/B-V format for 125 nuclides, and the report will be issued when the spontaneous fission data have been incorporated into the file.

Table 2.3

UK Chemical Nuclear Data Libraries Status Table - October, 1981

Data	Present Status	File Development
1. Fission product Decay Data	Exists as UKFPDD-2 (ENDF/B-IV format) - replaces UKFPDD-1 Total no. of nuclides = 855 Radioactive nuclides = 736 Ground state = 715 1st excited state = 133 2nd excited state = 5 Nuclides with spectra = 390 Total no. of gamma lines = 11,978 Total no. of beta$^-$ lines = 3592 Total no. of beta$^+$ lines = 91	Data acquisition for future revision
2. Activation Product Decay Data	Available in ENDF/B-IV format for 91 nuclides as UKPADD-1	Maintenance and improvements are in hand (Nichols, AEEW) for a library of 410 nuclides
3. Heavy Element and Actinide Decay Data	Exists as UKHEDD-1 (ENDF/B-V format)- Total no. of nuclides = 125 Ground state = 111 1st Metastable state = 13 2nd Metastable state = 1 Total no. of alpha lines = 767 Total no. of beta$^-$ lines = 527 Total no. of beta$^+$ lines = 39 Total no. of gamma lines = 3474 Total no. of discrete electrons = 6755 Total no. of X-rays = 381	Spontaneous fission data being evaluated (James, AEEW) - completion expected by December 1981
4. Fission Yields	Available in ENDF/B-IV format, based on Crouch's second round of adjustment (Crouch 3). 60 new compilations have been added. Yields to isomeric states have been calculated on basis of ENDF/B-IV ratios and are included	It is hoped to publish this evaluation soon.
5. Delayed Neutrons	Tomlinson data are still recommended	Delayed neutron emission probabilities and half-lives of precursors have been evaluated. Neutron spectra await evaluation
6. (α,n) cross sections	None in library	Data being evaluated by Barnes (BNFL)
7. Gamma spectra from (n,γ) reactions	None in library	Data being compiled by Davies (BNL)

A comprehensive reassessment has been made of the decay data needs for the activation product decay data file (UKPADD-1). Requirements for such data have been identified involving 410 nuclides and extensive revision of this file is planned.

Data for a future revision of this fission product decay data file (UKFPDD-1) are being collected (Tobias). In particular spectral data for 12 nuclides have been found where they were not previously available. The decay data files for activation products, heavy elements and fission products have been used to produce combined tables of gamma radiation ordered by nuclide and by energy.

The half-life of ^{239}Pu has been re-evaluated (Nichols and Glover).

2.5.2 ^{235}Np decay data evaluation (A.L. Nichols (AEE))

An evaluation has been made of the decay data for ^{235}Np, a nuclide that has become popular to monitor neptunium chemistry in the environment. This nuclide produces few suitable emissions for quantitative analysis. Transition intensity data for the electron and X-ray emissions associated with the electron capture decay mode are proposed.

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

All the programs have been converted to fast disk storage from card decks. However when used to calculate decay heat the consistent sets gave good agreement with experimentally determined values. There are some places where this is not so and no reason can be given yet[1].

---

(1) A. Tobias. Prog. in Nucl. Energy, 5, (1980), pp. 1-93.

3. REACTOR PHYSICS DIVISION, AEE, WINFRITH

(Division head: Dr. G.C. Campbell)

3.1 The UK Nuclear Data Library and allied topics (R.W. Smith and J.S. Story)

(a) A fully validated 1981 edition of the UKNDL which supercedes all previous editions of the library was distributed within the UK and to the NEA Data Bank during the period of this review.

(b) The evaluation of the resonance parameters of ^{58}Fe up to ~350 keV was completed and an internal report issued. These data are being used for re-analysis of the ^{58}Fe capture reaction rates measured relative to ^{249}Pu fission in various ZEBRA cores. Work has continued on the partially completed evaluations for ^{54}Fe and ^{56}Fe.

(c) A report including a detailed user guide, has been produced for SIGAR7 and will be distributed in early 1982. This code generates an energy point tabulation from resonance parameters and applies Doppler broadening.

3.2 AEE counting laboratory (W.H. Taylor, M.F. Murphy, M.R. March)

3.2.1 Beta and gamma decay power from fast fission

(a) Beta decay power

Measurements of the beta decay power from fragments from the fast fission of ^{239}Pu and ^{235}U have been made following an intermittent irradiation in ZEBRA over a period of 43 days during which the fissile deposits experienced a total neutron fluence of $\sim 10^{16}$. The results of these measurements were compared with values calculated using the FISPIN code with the older (UKFPDD-1 + CROUCH-2) and the latest (UKFPDD-2 + C3I) data sets. The best agreement was obtained using the latest set where the deviations of C/E from unity were within the total experimental error (which had components of $\simeq \pm 2.5\%$ systematic and $\pm$ 1 to 5% random). Past experimental results for a continuous 10^5 second irradiation[1], which were previously compared with values calculated using the UKFPDD-1 + CROUCH-2 data have now been compared with values calculated using the UKFPDD-2 + C3I data. The values obtained using either data set were within the total experimental errors.

(b) Gamma-ray decay power

Measurements of the total gamma-ray power from ^{239}Pu and ^{235}U fission fragments following the 43 day irradiation in ZEBRA were made using the large liquid scintillation tank at AERE; high resolution gamma-ray spectrum measurements were also made. Both types of measurement were made as a

function of cooling time. A comparison of the total power results with values calculated using FISPIN with the UKFPDD-2 + C3I data showed that the calculated values were in general ~8% lower than the measured values.

Absolute activities of the different isotopes at cooling periods in the range 10^3 to 10^7 seconds were evaluated from the measured spectra. These were compared with activity values calculated using FISPIN with the latest UKFPDD-2 + C3I data. This comparison showed that the calculated values were systematically lower than the measured values by ~12%.

3.2.2 Activation cross sections of fast reactor structural materials

The absolute activities induced in the components of fast reactor steels (normalised to the absolute ^{239}Pu fission rate) have been determined after the samples had been irradiated in the fissile and fertile regions of a range of ZEBRA assemblies. The results of comparisons between experiment and values calculated using fluxes from standard ZEBRA calculations with cross sections from the FD5 activation tape, have enabled bias factors to be provided for the more important contributors to the steel activations. The use of these factors will enable reaction rate ratios to be calculated with uncertainties of about ±10%.

3.2.3 Studies of the $^{93}Nb(n,n')^{93m}Nb$ reaction

Absolute determinations of the $^{93}Nb(n,n')^{93m}Nb$ reaction rates in EBRII (USA) and BR2 (Belgium) reactors have been made by six European Laboratories, including AEE Winfrith, as part of a niobium dosimetry intercomparison experiment. The estimated accuracy of these determinations was ≃ ± 3% and the conclusion (based on the spread of the results) was that the absolute reaction could be determined routinely with an accuracy of ≃ ± 6%. These results are to be presented in a joint paper to the ASTM/EURATOM Symposium on Dosimetry in Washington on March 22-26 1982.

A trial measurement of the differential cross section for this reaction has been made using the DYNAMITRON machine at Birmingham Radiation Centre (BRC). This has been done with the joint efforts of AERE, AEE and BRC and is reported earlier in 1.7.

3.2.4 Actinide fission rate measurements

Absolute fission rates of ^{235}U, ^{238}U, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu, ^{241}Am, ^{243}Am and ^{244}Cm have been determined in the fissile and fertile regions of a heterogeneous (annular) fast reactor core in ZEBRA. The random uncertainties on the measured values were in the range 0.1 to 1.0% and the systematic uncertainties in the range 0.6 to 9.4%. These values

have been compared with values calculated using standard methods. The C/E ratios in the core zone are very similar to those found in an earlier ZEBRA core. The recent Harwell evaluations for ^{241}Am and ^{243}Am give calculated ratios in much better agreement with experiment. The agreement for ^{244}Cm is poor. The Am and Cm fission rate predictions tend to be worse at the centre of the breeder island suggesting that the cross sections in lower energy groups are in error.

---

(1) M.F. Murphy, W.H. Taylor, D.W. Sweet and M.R. March. AEEW-R 1212 (1978).

3.3 Fission product decay heat (M.F. James)

See invited paper to Nuclear Data Forum on p. 8.

4. PHYSICS DEPARTMENT, NATIONAL RADIOLOGICAL PROTECTION BOARD

(Head of Department: J.A. Reissland)

4.1 Neutron cross section and kerma calculations from 20 to 50 MeV (P.J. Dimbylow)

Cross sections and kerma values have been calculated for C, N, O, Mg, Al, P, S, Ar and Ca from 20 to 50 MeV incident neutron energy. The optical model has been used to calculate total and elastic cross sections and a Haüser-Feshbach model with precompound corrections to calculate reaction cross sections. The direct reaction stimulation of low-lying levels produced by (n,n'), (n,p) and (n,d) reactions has also been investigated for C, N and O.

The distorted-waves Born-approximation code, DWUCK[1] has been converted for use on the IBM 3033 at Harwell. The levels at 4.43 and 9.64 MeV in ^{12}C, and 6.13 and 6.92 MeV in ^{16}O are known to undergo strong collective excitations. DWUCK has been used to calculate the direct reaction component of inelastic scattering to these levels with deformation parameters obtained mainly from reported analyses of (p,p') scattering. The direct component of (n,d) reactions has been calculated using the proton pick-up model. The (n,p) reactions were considered to be mediated by a combination of spin-exchange and isospin-exchange potentials. A set of cross sections and kerma values for C, N and O[2] which combine direct reaction and statistical model components has been prepared.

---

(1) P.D. Kunz, University of Colorado, Private Communication.

(2) P.J. Dimbylow, 'Neutron cross sections and kerma values for elements of biomedical importance' in Proceedings of the Fourth Neutron Dosimetry Conference, Munich 1981, p. 341, Vol. I.

5. DIVISION OF RADIATION SCIENCE AND ACOUSTICS

NATIONAL PHYSICAL LABORATORY

(Supt. Dr. W.A. Jennings)

5.1 Neutron source calibrations (E.J. Axton, A.G. Bardell)

Following the acquisition of high purity $MnSO_4$ olution for the manganese bath facility a series of measurements has been completed at ten different manganese concentrations with a RaBe photoneutron source in the 50 cm diameter bath and a ^{252}Cf source in the 100 cm diameter bath. Analysis of the results will yield values for the source strength and for the H/Mn thermal neutron capture cross section ratio which can be compared with historic values corrected for the effects of recently discovered chemical impurities in the old solution. All neutron source strengths calibrated at NPL prior to 1981 have been revalued upwards by 0.5%. A paper describing this work has been submitted to the journal Metrologia.

5.2 International comparison of neutron source strength (E.J. Axton, A.G. Bardell)

The comparison organised by the International Bureau of Weights and Measures[1] (BIPM) is in progress and measurements are expected to be completed soon. All laboratories have now completed their measurements with the exception of BARC and NBS. A list of participants is given below.

National Bureau of Standards (NBS) USA
Bureau International des Poids et Mêsures (BIPM)
Physikalisch-Technische Bundesanstalt (PTB) Federal Republic of Germany
Comitato Nazionale Energia Nucleare (CNEN) Italy
Centre d'Études Nucleaires (CEN) France
Laboratoire de Metrologie des Rayonnements Ionisants (LMRI) France
Amt fur Standardisierung, Messwesen und Warenprufung (ASMW) German Democratic Republic
V G Khlopin Radium Institute (VG KRI) USSR
D I Mendeleev Institute of Metrology (VN IIM) USSR
Electrotechnical Laboratory (ETL) Japan
Bhabha Atomic Research Centre (BARC) India
National Physical Laboratory (NPL) UK

---

(1) NPL represents the UK upon the various consultative committees of the Comité International des Poids et Mesures (CIPM) which guides the work of BIPM.

5.3 International comparison of neutron fluence rate organised by BIPM (J.B. Hunt, V.E. Lewis, T.B. Ryves)

Details of the neutron energies and transfer methods were given in UKNDC(80)P96, p.89. The current situation regarding the various legs of the comparison are described below.

(a) $^{115}In(n,n')^{115m}In$ comparison at 2.5, 5.0, 14.8 MeV.

Co-ordinator BCMN. 10 participants (including China).

Simultaneous measurements during 1981. Analysis mid-1982.

(b) Nb/Zr comparison at 14.8 MeV.

Co-ordinator NPL.9 participants (including China).

Simultaneous measurements during 1981. Analysis early in 1982.

(c) $^{115}In(n,\gamma)^{116m}In$ comparison at 144 and 565 keV.

Co-ordinator NPL. 6 participants so far.

Sequential measurements till end of 1983. Analysis 1984.

(d) ^{235}U fission chamber at all energies and ^{238}U fission chamber at 2.5, 5.0 and 14.8 MeV.

Co-ordinator AERE/NPL.8 participants so far (including linear accelerators).

Sequential measurements till end of 1984. Analysis 1985.

5.4 Evaluation of neutron cross sections at 14.7 MeV (J.G. Hayes, T.B. Ryves)

A paper has been published in Ann. of Nuc. Energy 8, 469 with the following abstract.

"A simultaneous evaluaton of the $^{27}Al(n,\alpha)$, $^{56}Fe(n,p)$, $^{63}Cu(n,2n)$, $^{65}Cu(n,2n)$, $^{197}Au(n,2n)$, $^{93}Nb(n,2n)$, $^{32}S(n,p)$ and $^{1}H(n,n)$ cross sections at an energy of 14.70 MeV has yielded a consistent set with uncertainties (1σ) of 0.6 - 2.8%. The large number of experimental activation results included were all considered as ratios between cross sections and the method of weighted least-squares was used in the evaluation, the results being normalised to the associated particle measurements.

The results have been compared with several other evaluations and are mostly in agreement within the errors. It was found that the ENDF/B5 ratio of the two most commonly used standard cross sections, $^{27}Al(n,\alpha)$ and $^{56}Fe(n,p)$, was 7.5% higher than our ratio. The $^{65}Cu(n,2n)$ activation cross section, which has often been used as a reference, cannot be determined to better than about 3% until the ^{64}Cu decay scheme is better characterised."

5.5 Nuclear decay scheme measurements (P. Christmas, D. Smith, M.J. Woods R.A. Mercer)

Half-life measurements were continued for a number of radionuclides including ^{3}H and $^{152,154}Eu$. As a result of work carried out in collaboration with the MRC Cyclotron Unit, a value of (270.82 ± 0.27) days has been published[1] for the half-life of ^{68}Ge.

NPL has contributed to a compilation of experimental decay data for radionuclides used in the efficiency calibration of γ-ray spectrometers, prepared by the International Committee for Radionuclide Metrology (ICRM) and to be published by the National Bureau of Standards (NBS).

(1) S.L. Waters, G.R. Forse, P.L. Horlock and M.J. Woods (1981). Int. J. Appl. Radiat. Isotopes, 32, 757.

6. BIRMINGHAM RADIATION CENTRE*, UNIVERSITY OF BIRMINGHAM

(Director: Professor J. Walker)

6.1 Neutron spectra from (α,n) sources (J.G. Owen, D.R. Weaver and J. Walker)

(Relevant to request number 31)

Last year (UKNDC(81)P100, p.80) we reported measurements made using our ^{3}He spectrometer of the neutron spectrum from the Harwell 100 mCi Am/Li source; a detailed paper has now been published[1]. As a consequence of this work the National Physical Laboratory agreed to lend us their 5 Ci Am/Li source for similar measurements to be made on it. It is known that the neutron emission from this source is anisotropic, (Bardell and Hunt, private communication) and the neutron spectrum was therefore measured at three different orientations. Because all three results were very similar, the pulse height distributions were combined and unfolded to give an average source spectrum which is shown in Fig. 6.1, where it is compared

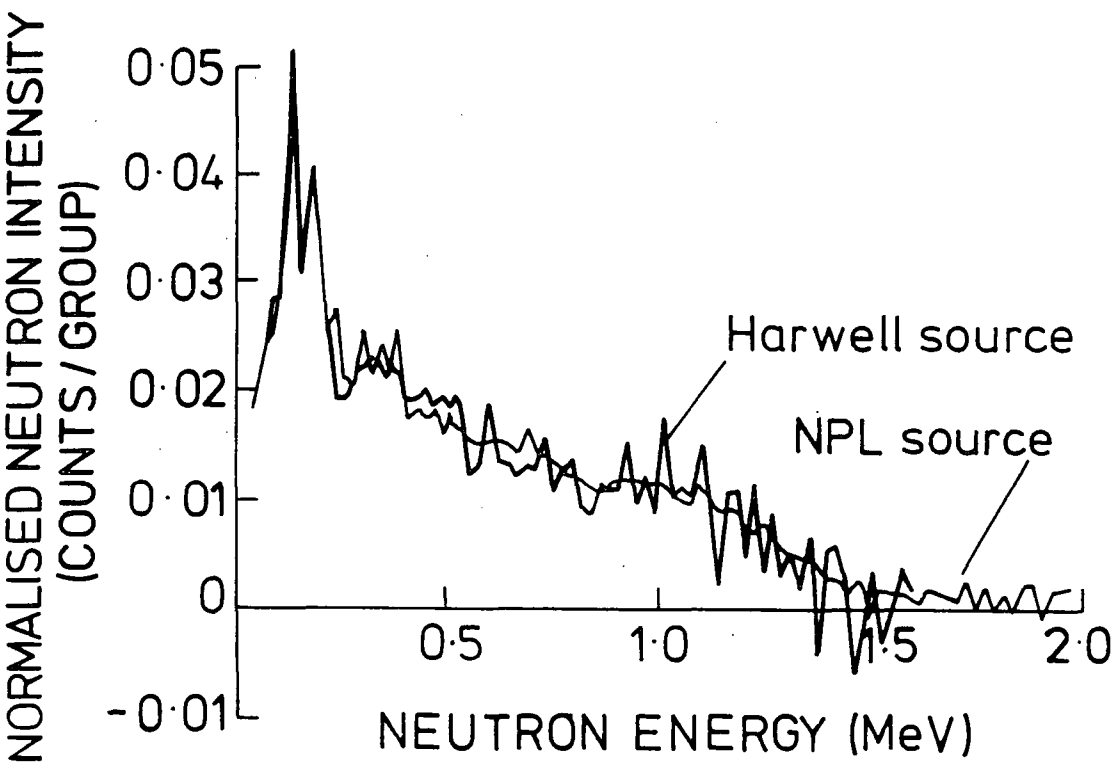

Fig. 6.1 The neutron spectrum from the Harwell and NPL Am/Li sources

*Operated jointly by the Universities of Birmingham and Aston in Birmingham.

with the spectrum from the Harwell source. Within the limits of error there is good agreement between the two data sets. Full covariance error matrices were calculated for all the spectra. The results of this measurement have been submitted for publication and numerical results are available from the authors.

A measurement is in progress of the neutron spectrum from an Am/F source, also on loan from NPL.

6.2 Delayed neutron energy spectra from ^{235}U and ^{239}Pu (J. Walker, D.R. Weaver and J.G. Owen)
(Relevant to request numbers 1184 and 1401)

Measurements in this programme have been resticted this year to the completion of the response function of our second ^{3}He spectrometer for neutron energies up to 2 MeV. In the data anlysis, the covariance method described in connection with the neutron spectrum from an Am/Li source[1] has been used with the delayed neutron data for ^{235}U and ^{239}Pu measured earlier on the Birmingham Dynamitron and the Harwell IBIS and Tandem accelerators (see UKNDC(81)P100, p. 81). So far only the results from our older Shalev-type ^{3}He counter have been analysed, and have shown the need for more information in the case of ^{239}Pu. In the first instance this is being obtained in two ways: (a) from the single-parameter (pulse height, no rise-time) data taken with the new counter during the IBIS experiments, now that the full response function of this counter has been measured; (b) by including $^{3}He(n,p)$ events in short rise-time channels. The rise-time analysis employed so far with our Shalev detector has meant that short rise-time pulses, although recorded, have been discarded before analysis because of contamination by pulses from ^{3}He recoils. A method for using them is now being investigated.

---

(1) J.G. Owen, D.R. Weaver and J. Walker. The calibration of a ^{3}He spectrometer and its use to measure the neutron spectrum from an Am/Li source. Nucl. Instr. Meth. 188 (1981), 579-593.

7. UNIVERSITY OF EDINBURGH, DEPARTMENT OF PHYSICS

7.1 The polarisation and differential cross section for the elastic scattering of 3 MeV neutrons by heavy nuclei (J.R.M. Annand and R.B. Galloway)

(Relevant to request numbers 1008 and 1213)

Simultaneous measurements of the angular dependence of polarisation due to elastic scattering and of the elastic differential cross section for 3 MeV neutrons scattered by W, Hg, Tl, Pb, Bi and U are being made to test the results of optical model and Haüser-Feshbach calculations and optimum optical model parameters are being sought.

The new polarimeter now in use is providing data of much improved accuracy compared with the best that could be obtained from the instrument used previously[1]. The new polarimeter provides simultaneous scattering asymmetry measurements at 11 scattering angles with computer controlled interchange of detectors to eliminate any instrumental asymmetry and removal of the scattering sample for background measurement. Measurements are made at two sets of angles differing by 7° to provide analysing power values at 22 angles from 20° to 167°, as shown in Fig. 7.1. Compared with the previous work[1] each distribution contains twice as many data points, each with an accuracy improved by a factor of about 5.

In the present instrument intercalibration of the detection efficiencies of the 22 detectors forms part of the computer controlled measurement sequence so that accurate differential cross section data can also be obtained. Fig. 7.2 illustrates a test on this aspect of the measurements in which two cylindrical iron scattering samples were used. The large sample measured 50 mm in diameter by 50 mm high and the small sample 25 mm in diameter by 50 mm high. Close agreement is shown between the differential cross sections deduced from the two samples after correction for flux attenuation, multiple scattering and finite geometry effects.

---

(1) Amena Begum and R.B. Galloway, J. Phys. G7 (1981), 535.

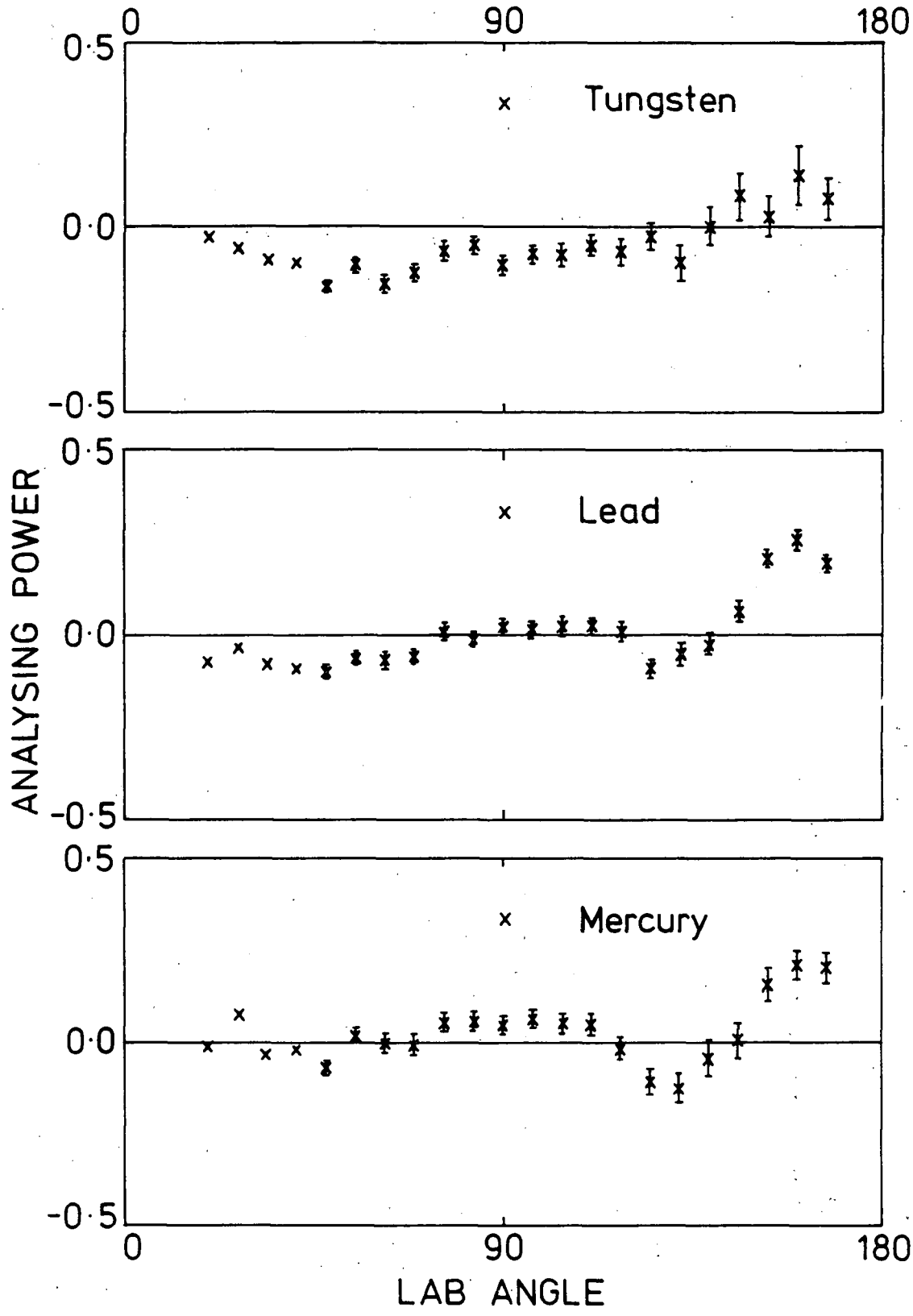

Fig. 7.1 Analysing power for 3 MeV neutrons as measured by the new polarimeter

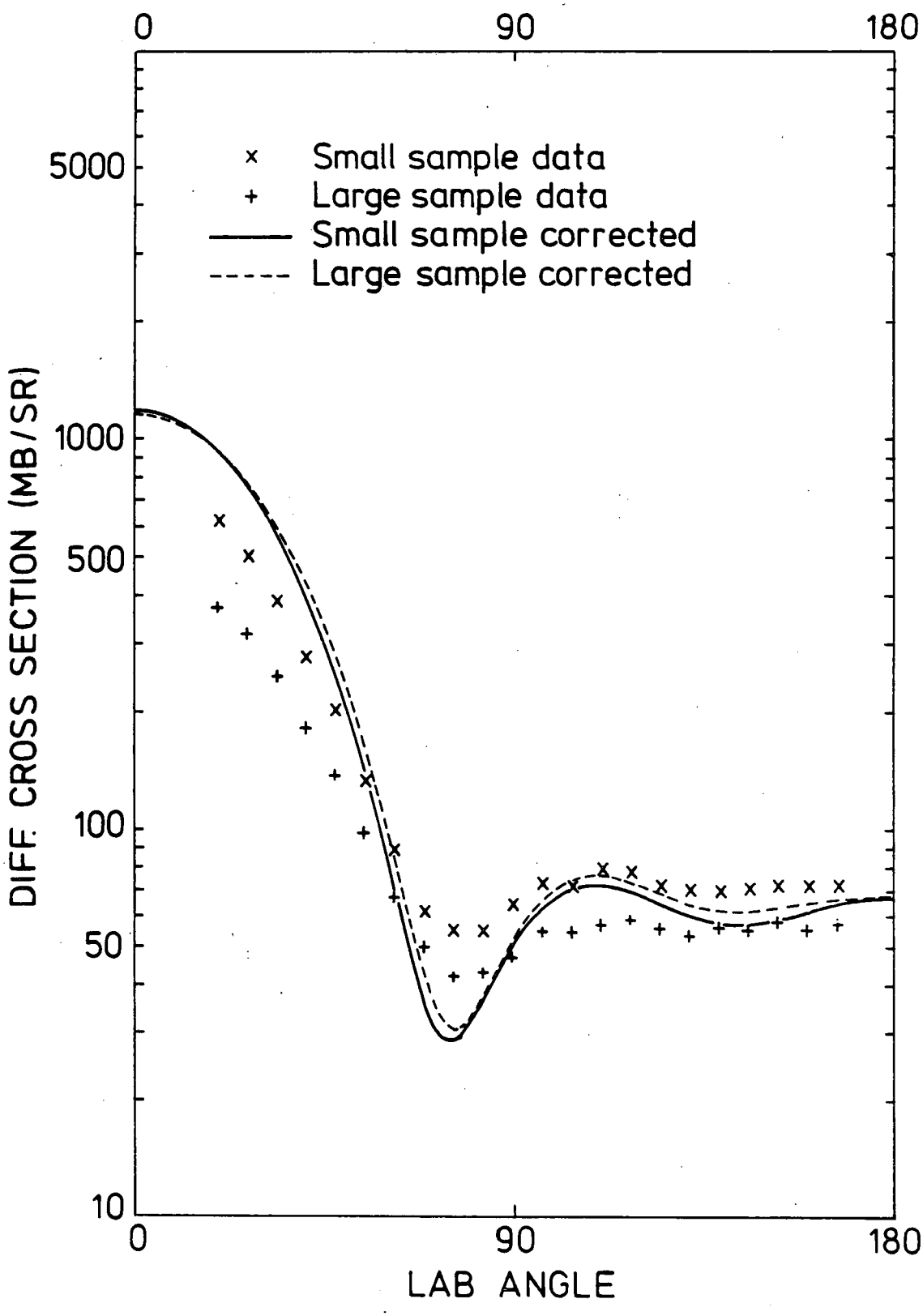

Fig. 7.2 Differential elastic cross section for 3 MeV neutrons incident upon iron

8. DEPARTMENT OF PHYSICS, UNIVERSITY OF BIRMINGHAM

8.1 Fast neutron spectrometry and its application in medical physics and fusion reactor data assessment (M.C. Scott, J.W. Bennett and J.S. Petler)

A miniature (~1.5 ml) NE 213 scintillator attached to a 23.5 cm quartz light guide has been used to make a series of absolute neutron spectrum measurements in a water filled anthropomorphic phantom irradiated by a point source of 14 MeV neutrons[1]. The spectra were unfolded for preliminary analysis using a differential code because the presence of the light guide altered the response function for the bare detector. It would appear that scintillator photon transport in the detector and light guide may have to be modelled, in addition to the neutron behaviour, to produce agreement with measurement.

Some of the results are shown in Fig. 8.1 where the effects of different intervening water thicknesses in softening the neutron spectrum can be seen clearly, as can the effect of enhanced leakage from limbs compared to the thorax. For convenience, the spectra are normalised over the uncollided 14 MeV neutron peak.

The study of the importance of charged particle production in the wall on hydrogen proportional counter response functions has now been completed. This involved modelling the (n,α), (n,p) and (n,d) reactions in the stainless steel walls and filling gas of a spherical counter and determining the track length distribution and hence pulse height for the different reaction products. The results of one such calculation for 14.2 MeV neutrons on a counter filled with a hydrogen argon mixture (2 atmos. H_2 and 8 atmos. Ar) are shown in Fig. 8.2, where it can be seen that at the upper operating limit for this detector, 2.5 MeV, protons from the wall contribute about 80% of the pulses. At the lower operating limit of around 600 keV, protons from the wall still contribute about one third of the total signal.

In Fig. 8.3 a comparison is shown between the calculated and measured neutron response function for the same detector, normalised arbitrarily over the lower end of the spectrum. Here we see that there is good agreement over the detector's operating range, 0.6 - 2.5 MeV, but that between 4 and 5 MeV and between 6 and 7 MeV there are discrepancies, the origin of which has not been resolved. It could be due to a nuclear data problem, or to uncertainties in the effect of space charge on gas multiplication for the more densely ionising α-particles. This problem is

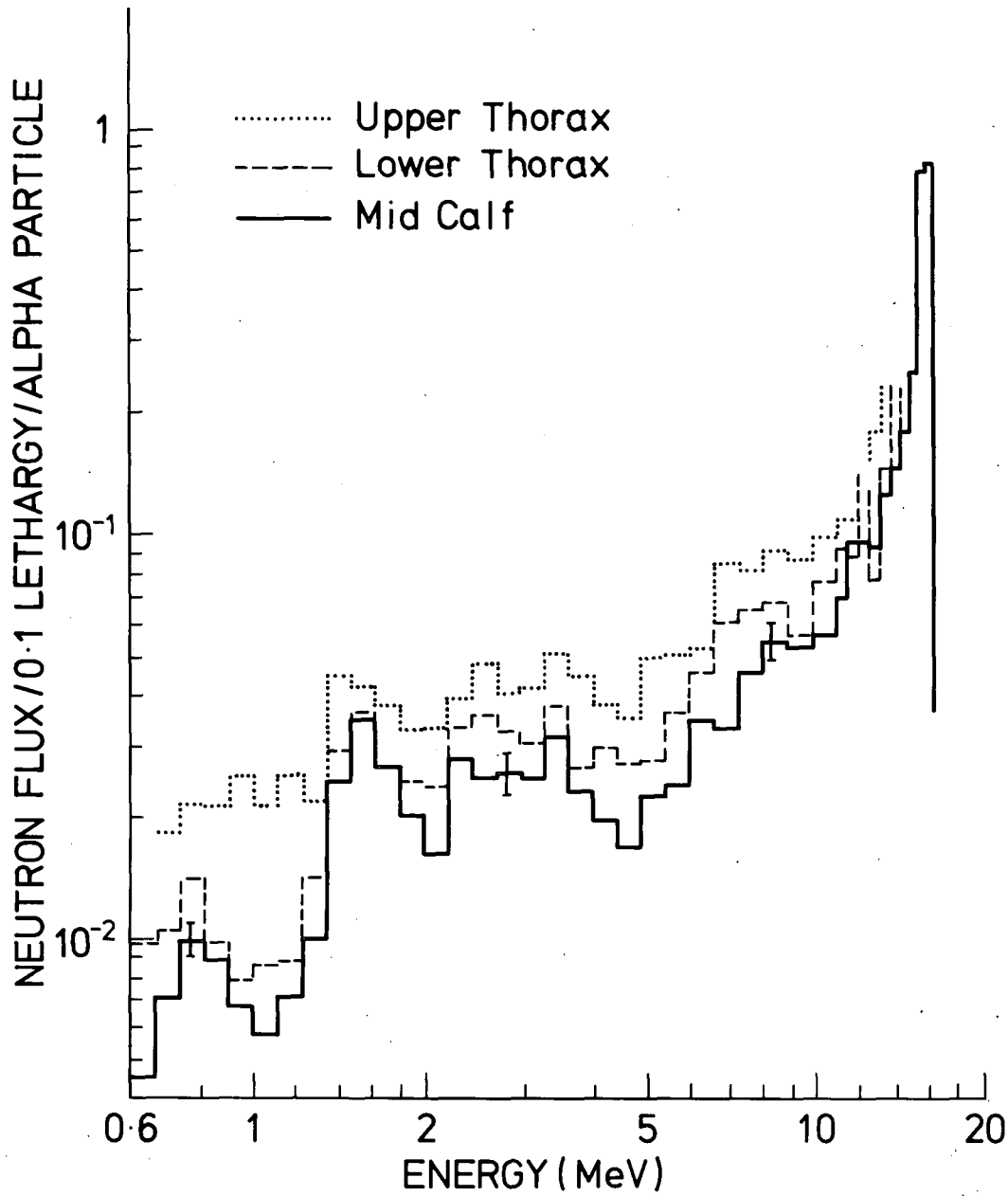

Fig. 8.1 Measured neutron spectrum in a water-filled anthropomorphic phantom, at lower and upper positions in the mid thorax, and at the centre of the lower leg.

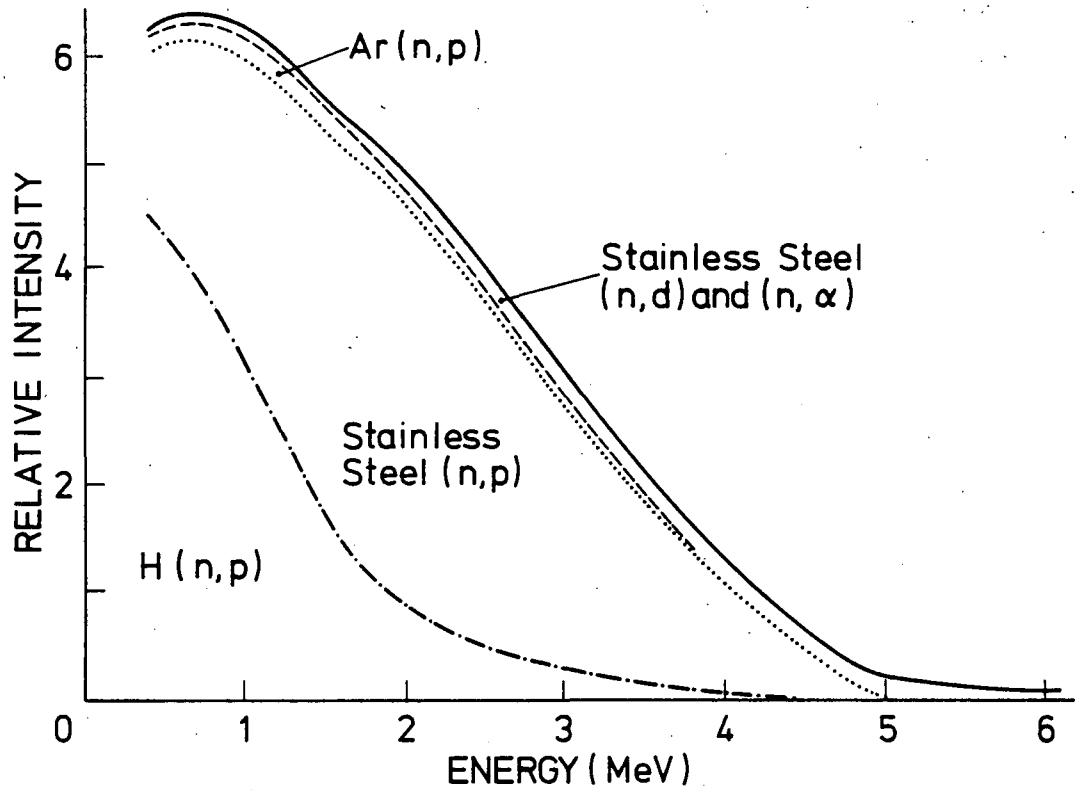

Fig. 8.2 Components of the response of a 4 cm diameter spherical proportional counter (2 atmos. H_2 and 8 atmos. Ar) exposed to 14.2 MeV neutrons.

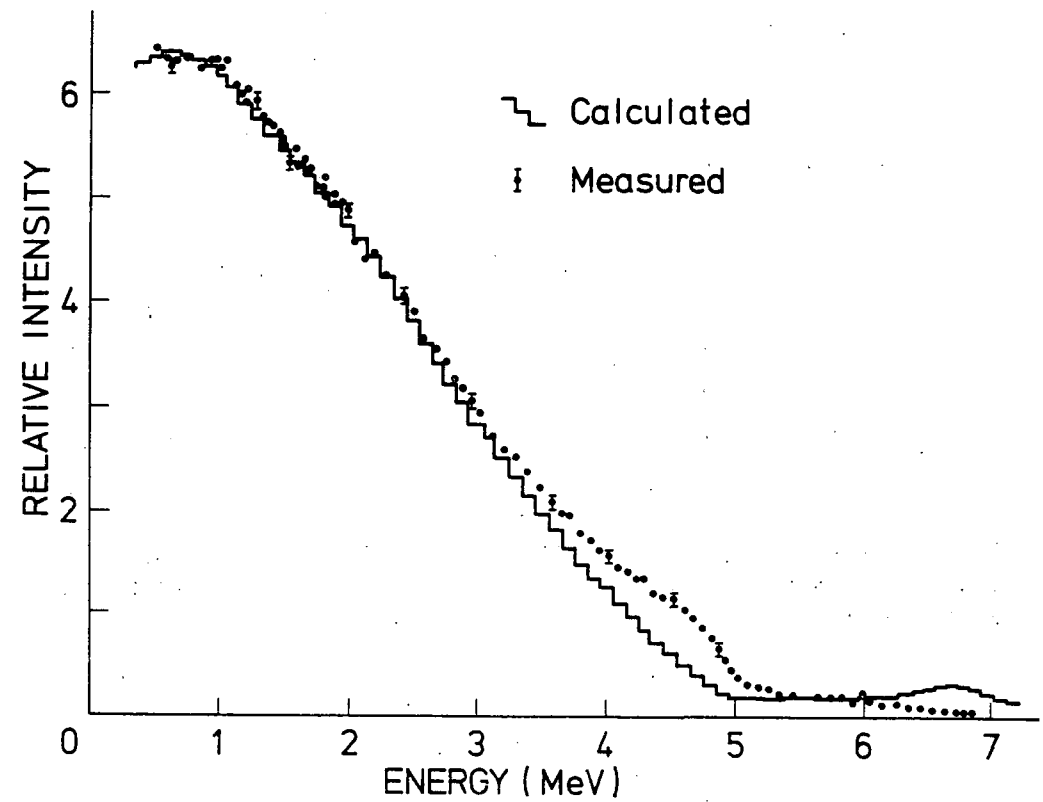

Fig. 8.3 Comparison of experimental and predicted detector response functions for 14.2 MeV neutrons (detector as in Fig. 8.2)

related to the more general one of non-linear gas multiplication in proportional counters[3], but it is very difficult to devise experiments which relate to the actual track length distribution and which enable one to measure the pulse height-ionisation characteristics for all the charged particles involved.

Since the existing analysis programs do not allow for other than proton recoils in hydrogen, these differences in response function will usually affect the unfolded spectra. In earlier work in lithium fluoride the measured spectra were in error at the high energy end, which we tentatively ascribed to the inadequate response functions[2]. It is encouraging to note that using the new response functions the unfolded spectrum is in better agreement at the upper energy end with both scintillation counter measurements and calculation. The magnitude of the changes in neutron spectrum from a spherical assembly of LiF having a certain D-T source can be seen in Fig. 8.4, the flux being increased by a factor of ~2 at 2 MeV. At lower energies the changes are much less.

Proportional counter measurements have also been made in the phantom mentioned earlier. In addition, a large gridded proportional counter (0.7 m high and 0.2 m diameter) has been built, the grid wires serving to define an inner and outer detector volume to eliminate wall-effects. This will be used both for studying leakage spectra and neutron spectra from bare targets and hopefully for studying charged particle producing reactions in the light elements of biological interest, N, O and C.

---

(1) M.C. Scott, J.W. Bennett and J.S. Petler, Proc. 4th Symp. on Neutron Dosimetry, EVR 7748, 2, (1981), 167-180, "Neutron Spectrum Studies in Phantoms from 30 MeV to 14 MeV".

(2) M.C. Scott, R. Kohi-Fayegh, N. Evans, L.J. Perkins and N.P. Taylor, Proc. NEA Specialists Meeting on Nuclear Data and Benchmarks for Reactor Shielding, Paris 1980, pp. 345-356, "Experimental Techniques for 14 MeV Neutron Benchmark Studies".

(3) I.R. Brearley, A. Bore, N. Evans and M.C. Scott, Nucl. Instr. Meth. (1982) In Press. "Some Aspects of Proportional Counter Behaviour for Neutron Spectrometry".

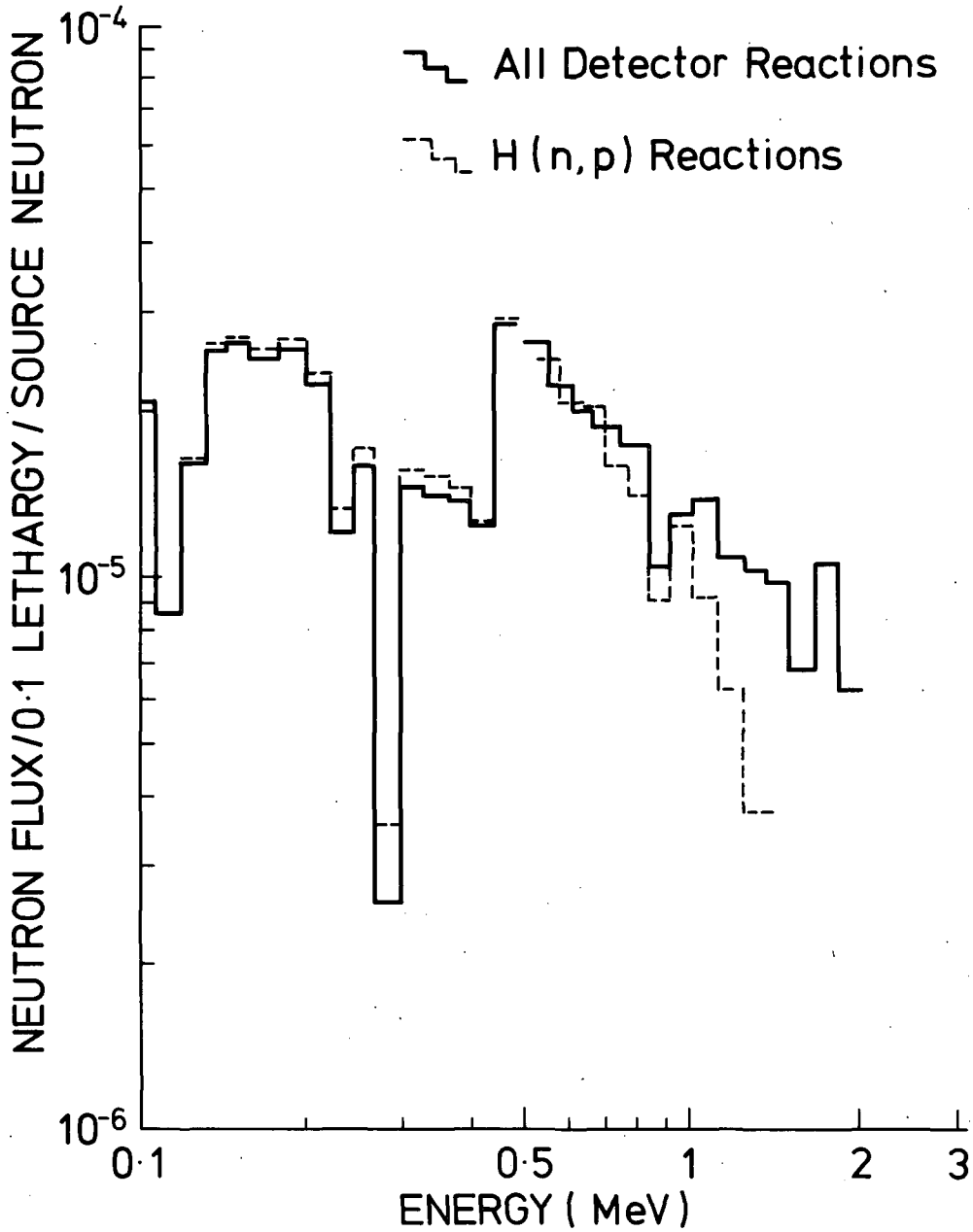

Fig. 8.4 Comparison of the neutron spectrum in LiF unfolded with and without allowing for charged particle production in the walls and in argon (detector as in Fig. 8.2)