

FISSION PRODUCT NUCLEAR DATA (FPND) Vol.1

PROCEEDINGS OF A PANEL ON FISSION PRODUCT NUCLEAR DATA ORGANIZED BY THE INTERNATIONAL ATOMIC ENERGY AGENCY AND HELD IN BOLOGNA, 26–30 NOVEMBER 1973



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FOREWORD

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

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

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

LIST OF ABBREVIATIONS

BWR	7	Boiling water reactor
CCDN	11 12	Centre de Compilation des Données Nucléaires NDCC = Neutron Data Compilation Center (of NEA)
EANDC	=	European-American Nuclear Data Committee
FP	=	Fission Product
FPND		Fission product nuclear data
FRO		Swedish zero power fast reactor
HTGR	#	High temperature gas cooled reactor
IAEA	Ħ	International Atomic Energy Agency
INDC	a	International Nuclear Data Committee
LMFBR	=	Liquid Metal fast breeder reactor
ND	13	Nuclear data
NDS	đ	Nuclear Data Section of the IAEA
NEA	-	Nuclear Energy Agency
ORELA	Ŧ	Oak Ridge linear accelerator
PFR		Prototype fast reactor
PWR	12	Pressurized water reactor
RP	=	Review paper
RPI	Ħ	Rensselaer Polytechnic Institute
WRENDA	=	World Request List for Neutron Data Measurements

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Panel on Fission Product Nuclear Data, Part 1:

REVIEW PAPERS AND OBSERVATIONS, CONCLUSIONS AND RECOMMENDATIONS OF THE PANEL

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Note:

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Scientific Secretaries: M. Lammer) Nuclear Data Section J.J. Schmidt) Div. of Research & Laboratories, IAEA

PROGRAMME

MONDAY, 26 NOVEMBER Morning Opening and Organization of the Panel Review 1 a M. Lammer Introductory talk IAEA Review 1 b S. Valente List of compilations. CCDN Saclay evaluations and computer codes of FPND (no oral presentation) SESSION I: FPND user needs in application fields Chairman: R.H. Flowers Review 3 J.G. Tyror Importance of FPND in the ARE Winfrith physics design of power reactor cores Discussion Review 4* C. Devillers Importance of FPND for CEN, Saclay engineering design and operation of reactors Report M.Ya. Kulakovsky USSR survey on topic of FEI, Obninsk review 4 Discussion Review 5 W.J. Maeck FPND requirements for Allied Chem. Corp. determination of nuclear fuel Idaho Falls burnup Discussion

Presented by M. Lott

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	Discussion		
Chairman:	E. Merz		
	Review 7	E. Merz KFA Juelich	Importance of FPND for fuel handling
	Discussion		
	Review 2	A.K. Ganguly BARC, Bombay	FPND and environmental aspects of the nuclear fuel cycle
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	Review 8	<u>W.K.G. Kuehn</u> E.G. Niemann Inst. f. Strahlen- botanik, Hannover	Importance of FPND in life sciences, agriculture and industrial technologies
	Review 9	E.L. Alpen Battelle Northwest	Use of FPND in life sciences
	Discussion		
	General discuss	sion on the international and dissemination of FPI	l cooperation in the exchange ND information.
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Chairman:	W.J. Maeck		
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	Discussion		
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	Review 16	<u>A.R. de L. Musgrove</u> J.L. Cook, G.D. Trimble AAEC Lucas Heights	Prediction of unmeasured fission product yields		
	Discussion				
Afternoon	Review lla	W.H. Walker AECL, Chalk River	Status of fission product yield data for thermal reactors		
	Discussion				
	Review llb	J.G. Cuninghame AERE, Harwell	Review of fission product yield data for fast neutron fission		
	Report	W.J. Maeck	New measurements of fast fission yields		
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Afternoon					
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	Review 12	G. Rudstam Studsvik	Status of decay data of fission products		
	Discussion				

	Review 13	S. Amiel Soreq Nucl. Res. Centre	Status of delayed neutron data
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Chairman:	V. Benzi		
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	Discussion		
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	Report and Discussion	L.P. Abagyan	USSR activities in the field of neutron cross-section evaluation
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Morning		Subgroups to draft cond (meeting in parallel a	clusions and recommendations t the Centro di Calcolo)
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Chairman:	J.J. Schmidt		

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INTRODUCTION

M. Lammer

Nuclear Data Section, Division of Research and Laboratories

IAEA, Vienna, Austria

It is the purpose of this introduction to review briefly the historical development which led to the present meeting.

The original idea to convene a specialist meeting on the subject "Fission Product Nuclear Data" (FPND) was borne by Professor Benzi and Dr. Walker at the occasion of the Second IAEA Conference on Nuclear Data for Reactors at Helsinki, 1970. Recognizing the importance and timeliness of this subject the Agency took up this suggestion and envisaged to convene such a specialist meeting in 1973 provided that enough interest of Member States could be found. At its Fifth Meeting in 1972 the International Nuclear Data Committee (INDC) endorsed the Agency's plan and in the second half of 1972 the Nuclear Data Section (NDS) sent a circular letter to more than 200 users and producers of FPND soliciting their opinion and detailed comments on such a meeting. ' More than 100 replies were received, often representing collected opinions of several scientists or groups. The replies demonstrated a high interest of both FPND users and producers and provided important background information to a preparatory consultants meeting which the Agency convened in December 1972 at its headquarters in Vienna.

This meeting was attended by the following scientists:

Dr. R.H. Flowers from AERE Harwell, UK, representing FPND users; Prof. V. Benzi from the Centro di Calcolo, Bologna, Italy, representing FP microscopic data;

- Dr. M. Bustraan from RCN Petten, Netherlands, representing integral FPND measurements;
- Dr. S. Valente from NEA/CCDN Saclay, representing compilations and evaluations.

Apart from the scientific secretary, Mr. M. Lammer, and other members of NDS the meeting was also attended by other Agency members representing FPND interest in the fields of nuclear safety and environmental protection, reactors, life sciences, safeguards, INIS and physics.

in a se contacunto

The main task of the meeting was to develop a programme for the FPND Panel Meeting and to suggest appropriate ways for its preparation. It agreed on the following main objectives of the Panel:

- The panel should bring together users and producers of FPND.
- Users of FPND should specify their nuclear data requirements and their priorities in detail as a prerequisite of the panel's discussions, conclusions and recommendations.
- The status of knowledge of microscopic FPND should be reviewed together with a critical comparison of existing evaluations and compilations.
- The testing of microscopic FPND by integral measurements should be reviewed.
- The panel should identify and discuss further measurements, compilations and evaluations required to satisfy the needs of FPND users. It should aim at specific recommendations and measures for coordination of future work.

In order to meet these objectives and at the same time cover the discussion topics adequately, the preparatory meeting recommended the following organization of the Panel:

- a. The body of the Panel should be formed by 16 comprehensive review papers covering the full scope of use, status and testing of FPND and forming the basis for the discussions of the panel. In order to use the available meeting time most economically individual contributions instead of the reported separately should be included in the review papers with the additional benefit that the review papers represent the international state of the art in each topic.
- b. In order to achieve maximum efficiency the Panel, further to the review papers, should devote its time to discussions on open questions and to deriving recommendations for further work.
- c. The panel should discuss FPND of the following catagories most important for practical applications:
 - yields;
 - neutron cross sections;
 - decay data;
 - delayed neutron data

Because of the scope of the FPND to be discussed within one week, no FP data other than specified above should be discussed at this Panel. The Panel should therefore leave out problems and results of fundamental fission theory and experiments, which were dealt with by the Agency's Third Symposium on the Physics and Chemistry of Fission held at Rochester in the United States in August 1973. By the same token other data of importance to the nuclear fuel cycle which were mentioned in several answers to the aforementioned circullar letter should be excluded from the Panel's scope such as transactinium isotope production data, data on physical and chemical states of FP and others.

d. In accord with user needs, fission products of the following fissile and fertile muclides should receive first priority: Th-232, U-233, U-235, U-238, Pu-239, Pu-240 and Pu-241. Second priority should have fission products of the following muclides: all other more important heavy isotopes occurring in nuclear fuel cycles and more important spontaneously fissioning isotopes such as Cf-252.

Having fixed these main objectives and organization of the Panel, the Preparatory meeting then developed the programme for this Panel and defined the titles and contents of the review papers.

Review Paper No. 1b

LIST OF COMPILATIONS, EVALUATIONS AND COMPUTER CODES OF F.P.N.D.

S. Valente OECD/NEA/CCDN B.P.9 91190-Gif-sur-Yvette France

Abstract

The paper lists the compilations, evaluations and computer codes available at the time of the Fission Product Nuclear Data Panel (Bologna, November, 1973). It covers fission product yields, neutron reaction cross-sections, half lives, decay schemes, delayed neutron data and gamma-ray spectra.

- 1. Fission product yields (independent and cumulative) as a function of incident neutron energy or incident neutron spectrum, including binary and ternary yields.
- 2. Neutron reaction cross-sections of fission products as a function of neutron energy, in the following sequence of priority :
 - (a) Capture : sigma (E) and resonance integrals (including also capture cross-sections for destruction of long-lived fission products such as Sr-90, 1-129);
 - (b) Inelastic scattering ;
 - (c) Elastic scattering ;

- (d) Others such as neutron and gamma-production cross-sections.
- 3. Half-life data for fission product ground and metastable states.
- 4. Decay schemes, including :
 - (a) y-ray and conversion electron energies and intensities;
 - (b) β mean energies and intensities ;
 - (c) Decay branching ratios ;
 - (d) The total $\beta + \gamma$ energy yield.
- 5. Delayed neutron data as a function of incident neutron energy, including :
 - (a) Yields, half-lives energy spectra of delayed neutrons and average data for delayed neutron groups;
 - (b) Yields, half-lives, and emission probabilities of individual delayed-neutron precursors.
- 6. γ -ray spectra of fission products from neutron reactions (capture, inelastic scattering, etc.) as a function of incident neutron energy.
- 7. Computer codes for processing fission product data.

NOTE : The data types 1, 2(a), 3 and 4(c) are basic for fuel inventory and are referred to in working papers as 'Inventory data'.

1. Fission product yields

S. Katcoff	USA	Nucleonics 18/11(1960)201	1960	out of date
R.L. Ferguson et al.	USA	ORNL-3305	1962	out of date
Yu. A. Zysin et al.	USSR	Fiss. Prod. Yields and their mass distribution – Consultants Bureau. New York 1964	1964	out of date
C.A. Anderson	USA	LA-3383	1965	out of date, includes interpo- lated and calculated yields
I.F. Croall	UK	AERE-R-5086	1967	out of date
G. Cenacchi	Italy	(RT/FIMA(68)4 (CEC(68)6	1968 1968	difficult to obtain
E.A.C. Crouch	UK	AERE-R-6056	1969	purely calculated yields (fractional independent yields)
J. Bessis et al.	France	CEA-N-1180(1), (2), (3)	1969	difficult to obtain
A.C. Wahl	USA	SM-122/116	1969	only U-235 thermal yields, independent yields evaluated, cumulative yields are selected values but not evaluated
H.R. Von Gunten	Switzer.	Actinides Rev. 1.(1969)275	1969	

P. Hofmann	FRG	KFK-EXT-BER-6/70-2	1970	difficult to obtain
E.A.C. Crouch	UK	AERE-R-6642	1970	
K.F. Flynn et al.	USA	ANL-7749	1970	not freshly evaluated from original experimental data
D.R. Mathews et al.	USA	GULF-GA-B-12071	1971	
W.H. Walker	Canada	(CRRP-913 (CN-26/3 (AECL-3037 Part 11 (IAEA/SM-170/34	1960 1970 1972 1973	thermal yields only
M.E. Meek et al.	USA	(GEAP-5356 (GEAP-5505 (APED-5398 A (NEDO-12154	1967 1967 1968 197 2	compilation and evaluation
E.W. Sidebotham	UK	TRG -2143(R)	1972	purely calculated yields: yields were taken from other evaluations and readjusted by calculation
E.A.C. Crouch	UK	AERE-R-7207 IAEA/SM-170/94	1972 1973	compilation and evaluation (thermal and pile yields)
E.A.C. Crouch	UK	AERE-R-7209	1973	

C. Devillers	France	IAEA/SM-170/63	1973	
M. Lammer, et al.	Austria	IAEA/SM-170/13	1973	only evaluated data
E.A.C. Crouch	UK	AERE-R-7394	1973	fast yields

G.D. Joanou et al.	USA	(GA-2451, vols. 1,2,3 (GA-4132 (GA-4265	1962 1963 1963	
J.R. Stehn et al.	USA	BNL-325 2nd edition sup.2	1965/66	
G. Cenacchi	Italy	(RT/FIMA(68)4 (CEC(68)6	1968 1968	
J. Bessis et al.	France	CEA-N-1180(1), (2), (3)	1969	
H.D. Ferguson	Australia	AAEC/TM-520	1969	Description of GUNIA code
A.R. de L. Musgrove	Australia	(AAEC/E198 (AAEC/E198-1	1969 1970	description of computer library only (contains also resonance para- meters at 30 KeV).
Y.Liu	FRG	Juel-678 RG	1970	graphs of cross-section versus neutron energy calculated from exp. and eval. data
J.L. Cook	Australia	AAEC/TM 549	1970	description of computer libraries only
E.K. Rose	Australia	AAEC/TM 587	1971	description of computer libraries only
W.K. Bertram et al.	Australia	AAEC/E-214	1971	group cross-sections
D.R. Mathews et al.	USA	GULF-GA-B-12071	1971	

2. Neutron reaction cross-sections of fission products

A.Z. Nagy et al.	Hungary	Journ. Rad. An. Chem. <u>7</u> (1971) 365	1971	
V. Benzi et al.	ltaly	(CEC(7)9 (RT/F1(72)6	1971 1972	
L. Stewart et al.	USA	(LA-4901 (LA-4918	1972 1972	((for gamma-ray heating)
V. Benzi et al.	Italy	Bologna library	1972	supplementing australian library above 10 KeV. Data available at CCDN(Saclay), NNCSC(BNL) and NDS(Vienne)
NNCSC	USA	(ENDF/B-111 (ENDF/B-1V	1972 1974	data available at CCDN(Saclay) and NNCSC(BNL)
AAEC	Australia	Australian fiss. prod. cross- sect. library in UK format	1972	data available at CCDN(Saclay), NNCSC(BNL) and NDS(Vienne)
S. lijima et al.	Japan (JNDC- JAERI)	Unpublished	1972	
E. Clayton	Australia	AAEC/TM-619	1972	tabulated thermal cross-sections and resonance integrals calculated from the australian library of evaluated fission product point cross-sections.
F.W. Walker et al.	USA	11th Edition of the GE–NR Chart	1972	

W.H. Walker	Canada	AECL-3037 Part 1	1972	thermal, resonance integrals, pile (includes all exp. data)
J. Krebs, P. Ribon	France (CEN Saclay)	Unpublished	1973	
H. Matsunobu	Japan	Panel of F.P.N.D., Bologna, 26–30.11.73	1965- 1973	
C. Devillers	France	IAEA/SM-170/63	1973	description of computer library. Data available at CCDN (Saclay), NNCSC(BNL), NDS (Vienne).
O. Eder, M. Lammer	Austria	IAEA/SM-170/12	1973	freshly evaluated, updated and estimated cross-sections
W. Seelmann-Eggeber et al.	FRG	NUKLIDKARTE 4th edition	1973	available only in March, 1974
G. Lautenbach	Nether- lands	RCN-191	1973	

M.R. lyer et al.	India	AEET/HP/TH-16	1964	tabulations of selected data from other evaluations/superseded data.
J. Lindskog et al.		Alpha, Beta and Gamma- ray Spectroscopy, vol.11 P.1599. North-Holland, Amsterdam	1965	
R.J. Bullock et al.	UK	AERE-M-2058	1968	
T.R. England	USA	Thesis for Ph.D. at University of Wisconsin	1969	
N.R. Large et al.	UK	Nuclear Data Tables <u>A7</u> (1970) 477	1970	supersede R.J.Bullock et al. AERE-M-2058
Y. Liu	FRG	JUEL-678-RG	1970	selected from other evaluations
A.Z. Nagy et al.	Hungary	Journ . Rad . An . Chem . 7 (1971) 365	1971	
D.R. Mathews et al.	USA	GULF-GA-B-12071	1971	
A.H. Wa pstra et al.	Nether . Iands	The 1971 Atomic Mass Evaluation, Nuclear Data A9(1971) 265	1971	
C. Meixner	FRG	JUEL-813-RX	1971	source of data cf.JUEL-811-RX

3. Half-life data for fission product ground and metastable states.

F.W. Walker et al.	USA	llth Edition of the GE–NR Chart	1972	
Q.J. Eder, M. Lammer	Austria	IAEA/SM-170/12	1973	
A. Tobias	UK	CEG B-RD/B/M-2669	1973	recommended values after survey of original publications
W. Seelmann- Eggebert et al.	FRG	(Nuklidkarte (Nuklidkarte 4th Edition	1968 1973	available in March , 1974
D.J. Horen et al.	USA	Nuclear Data B Nuclear Data Sheets	Con- tinuing	
M.J. Martin	USA	ORNL-4923	1973	

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4. Decay schemes

Landolt-Börnstein		Energy Levels of Nuclei : A = 5 to A = 257, edited by A.M. Hellwege and K.H. Hellwege	1961	
B.S. Dzhelepov et al.	USSR	Decay Schemes of Radio- active Nuclei, Academy of Science Press, Moscow, 1958. English translation Pergamon Press, New York, 1961	1961	
B.S. Dzhelepov et al.	USSR	USSR Academy of Science Press, Moscow, 1963	1963	
R.L. Heath	USA	1DO-16880-1-2 TID-4500, 31st edition	1964	
C.M. Lederer et al.	USA	Tables of Isotopes	1967	
E.W. Sidebotham	UK	FYAWP(68) P.11	1968	
G. Cenacchi	Italy	(RT/FIMA(68)4 (CEC(68)6	1968 1968	
R. De Tourreil	France	CEA-N-1023	1968	
B. Barré et al.	France	CEA-N-1191, 1193	1969	
T.R. England	USA	Thesis for Ph.D. at University of Wisconsin	1969	(for decay heating)

I.P. Selinov	USSR	Izdatelstvo "Nauka", Moscow, 1970	1970	
M.J. Martin et al.	USA	Nuclear Data <u>8A</u> 1-2 1970	1970	
G.K. Crocker	USA	AD-659980	1970	
D.J. Horen et al	USA	ORNL-4627 ORNL-4730	1970 1971	pure levels schemes without indicating transitions
A.Z. Nagy et al.	Hungary	Journ. Rad. An. Chem. <u>7</u> (1971) 365	1971	
D.R. Marr et al.	USA	HEDL-TME-71-27	1971	tabulations containing not original evaluated data
C. Meixner	FRG	JUEL-811,812,813-RX	1971	
L. Costa et al.	France	(J. Nucl. En.25(1971)285 (J. Nucl. En.26(1972)431	1971 1972	
A.H. Wapstra et al.	Nether- Iands	The 1971 Atomic Mass Evaluation, Nuclear Data A9(1971)265	1971	
M.A. Wakat	USA	Nuclear Data <u>8A</u> (5-6)	1971	
J. Blachot et al.	France	(CEA-N-1290 (CEA-N-1526	1971 1972	

F.W. Walker et al.	USA	11th Edition of the GE-NR Chart	1972	
M. Sakai	Japan	Nuclear Data Table A10 (1972)511	1972	collective bands of levels for even-even nuclei
J. Mantel	USA	Int. J. Appl. Radiation lsot.23(1972)407	1972	
J. Krebs, P. Ribon	France (CEN Saclay)	Unpublished	1973	
C. Devillers et al.	France	IAEA/SM-170/63	1973	data available at CCDN(Saclay), NNCSC(BNL) and NDS(Vienne)
O.J. Eder and M. Lammer	Austría	IAEA/SM-170/12	1973	data available as printout from SGAE (Seibersdorf)Status 1971
A. Tobias	UK	(CEGB-RD/B/M1551 (CEGB-RD/B/M2058 (CEGB-RD/B/M2453 (CEGB-RD/B/M2356 (CEGB-RD/B/M2669	1970 1971 1972 1972 1973	
W. Seelmann-Eggebert et al.	FRG	(Nuklidkarte (Nuklidkarte 4th edition	1968 1973	available in March 1974
D.J. Horen et al.	USA	Nuclear Data B Nuclear Data Sheets	Conti- nuing	

R.J. Bullock et al.	UK	AERE-R-7213	1973	not yet published
R.L. Heath	USA (Idaho)	AEC Catalog of Gamma Rays of Fission Product Data	Con- tinuing	contains pure decay gamms spectra
V. Sangiust et al. Pol	ltaly (Milan ytechnic)	Gamma spectra for Fission products (short lived)	ln pre- paration	
M.J. Martin	USA	ORNL-4923	1973	

G.R. Keepin	USA	Physics of Kinetics Chapter IV. Addison- Wesley, Reading, Mass.	1965
P, Del Marmol	Belgium	Nuclear Data Tables <u>A6</u> P.141	1969
S. Amiel	lsra e l	Proceedings of Second Symposium on Physics & Chemistry of Fission, IAEA, Vienna(1969), P.569	1969
A. Evans et al.	USA	LA-DC-72-561	1972 (Conti- nuing)
L. Tomlinson	UK	AERE-R-6993	1972
F. Manero et al.	IAEA	Atomic Energy Review, vol.10, N°4, P.637	1972

5. Delayed neutron data as a function of incident neutron energy

Y-ray spectra of fission pro	baucts from	neutron reactions		
E. Der Mateosian et al.	USA	BNL-605(T-177)	1960	
G.A. Bartholomew et al.	Canada	(Nuclear Data A3 no.4,5,6 (Nuclear Data A5 no.1,2	1967 1968	tables and graphs of capture gammas.
R, Gunnink et al.	USA	UCID-CMM-1969	1969	lab. Livermore
L.V. Groshev et al.	USSR	Nuclear Data A5 no.3,4	1969	tables and graphs of capture gammas
J.B. Wilhelmy	USA	Dissertation, Univ. of California, Berkeley	1969	High-resolution gamma and X-ray spectroscopy on unseparated fission products.
R.H. Filby et al.	USA	Washington State Univ. report WSUNRC-97(2)	1970	
K.H. Appelman et al.	Holland	(RCN-72-004 (RCN-72-005 (RCN-72-037 (RCN-72-038 (RCN-72-123	1972 1972 1972 1972 1972	gamma spectra of fission products ; internal laboratory reports
J.R. Bird et al.	Australia	Nuclear Data All no.6	1973	tables and graphs of capture gammas
J.S. Story	UK (Winfrith)	UKAEA Library	Con- tinuing	ł

7. Fission product codes

AIMFIRE	Atomics International Multicycle Fuel Inventory Reprocessing Economics. Developed for estimation of fuel cycle costs. (Atomics International)
CINDER	A one-point depletion and fission product programme, WAPD-TM-334, August 1962, rev.1964. (Westinghouse Atomic Power Division)
CURIE	Fission product inventory code developed for evaluating its radiological hazards. (Atomics International)
FISP	A comprehensive computer programme for genera- ting fission product inventories. (BNL/NNCSC)
FISP 2	A programme for the calculation of fission product inventories, by R.H. Clarke and R.E. Utting, RD/B/N-1737, RD/B/N-1427.
FISP 4 and HYLAS 2	Updated versions of the computer programs for calculating radioactive fuel inventories, by S.M. Beynon, April 1973, CEGB Report RD/B/N-2633.
FISPRO II	A programme for the calculation of fast neutron radiative capture cross-sections of fission products, RT/FI(69)44. (CNEN, Bologna, Italy)
FISSPROD	G-20 computer program for calculation of fission product absorption in thermal reactors, by F. Lane AECL-3038 (1969) Chalk River, Canada.
FP-DP	A programme for the calculation of <u>decay power</u> of fission products, by K. Tasaka and N. Sasamoto.
F P- y	A programme for the calculation of gamma-ray spectrum of fission products, N. Sasamoto and K. Tasaka.
FPIC	Fission Product Inventory Code. Developed for heating and shielding analysis. (Lockheed Aircraft Corporation)

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FP-S	A programme for the calculation of fission product inventories, by K. Tasaka and N. Sasamoto, JAERI 1198(1971).
GABS	A code for treatment of fission products in burn up calculations, using recent nuclear data applied to graphite reactors, by Yosen Liu, JUL-678-RG(1970).
HYLAS	A programme for the calculation of fission product inventories, by R.H. Clarke : RD/B/N 1722 heavy metals.
ICON	A programme for the calculation of fission product inventories, by R.L. Faircloth and M.J. Hopper, AERE-R-6242 (1970).
ISOGEN	A computer code for radio <u>ISO</u> tope <u>GENeration</u> calculations, (General Electric, GE-HW)
KASCO	A programme to calculate heavy metal burn up and fission product yield in HTR, by R. Wagemann and D. Wahl, AVR-Report T -1, H5-X1 (1972).
ORIGEN	Oak Ridge Isotope GENeration. Originated as an FBR-fuel code. (ORNL)
PEPIN LE BREF PLUS PEPIN	A programme for the calculation of the activity of fission products, by R. de Tourreil, CEA-N-824 (October, 1967).
PHOEBE	A computer code for <u>PHoton Energy</u> and <u>Beta Energy</u> calculations : bulk radioactive and thermal properties of highly-enriched LWR-fuel. (ORNL)
PICFEE II	A programme for the calculation of the concentrations and the activity of fission products, C. Devillers et al., SERMA 1185 (January, 1973).

RIBD	Radioactive Isotope Buildup and Decay. Developed to evaluate biological hazards and heating effects accompanying radioactive decay. (United Nuclear)
SFP	A computer code for estimation of Spent Fuel Properties which influence reprocessing. (ORNL)
SOSUM	SOurces and SUM, for shielding and heat transfer at reactor fuel discharge. (Atomics International).

FISSION PRODUCT NUCLEAR DATA (FPND) AND ENVIRONMENTAL ASPECTS OF NUCLEAR FUEL CYCLE

by

M.R.Iyer, D.N.Sharma & <u>A.K. Ganguly</u> Bhabha Atomic Research Centre Health Physics Division Bombay 400085, India

ABSTRACT

The paper reviews the availability and incompleteness of FPND that are important for the evaluation of environmental impact of release of fiesion products from various operations in the nuclear fuel cycle. The fields where relevance of FPND in environmental investigation and safety are well established are listed. It is emphasized that in every environmental and regional setting the investigator is required to discover for himself the significance of the presence of specific radio-nuclides in their respective context. Possible global environmental consequences of fission product releases are indicated as may arise from H-3, Kr-85, unfissioned fissile matter and neutron emission in nuclear explosions and fuel reprocessing. It is observed that date on independent yield in spontaneous fission or fission induced by fast neutron are mostly not available. There is a dearth of information on radiation from fission products at short times (upto a few seconds) after fission. The data on fission products are mostly better known only for three or four products next to the stable one in the chain. As predicted by a model study, out of a total of 671 fission products in the mass range of 80 to 156, approximately 333 remain

to be identified. Dependence of termary fission yields specially of H-3 on neutron energy is not well established. With the build up of nuclides of low spontaneous fission half-lives in the reactors and presence of these in waste streams from processing plants, precise FPND data and neutron cross section data including multiple neutron capture cross sections assume significance. Importance of investigations on recoil chemistry of fission products and transuranic elements and auger shower that determines the chemical state of the nuclear species in the environment, has been emphasised. Further, the paper discusses different possibilities of getting such information and emphasises the need for refinement of systematics and theoretical models for predicting FPND which are difficult to obtain directly through experiments.

1. INTRODUCTION:

In the present review we will draw a distinction between Fission Fragments (FF) - as those nuclear species that are formed immediately after fission and Fission Products (FP) - as those nuclear species resulting after prompt neutron evaporation from the fragments. Fig.l shows the limits of the neutron number of FF for each Z formed in the neutron induced fission of U-235 (1.7). Similar figures could be drawn for other fissioning nuclides also.

1.1 Release of Heavy Elements from Processing of Fissile and Fertile Materials

The nuclear fuel cycle starts with the mining and milling of uranium and thorium ores and consideration of environmental aspects has to take into account the release to the environment, of long-lived naturally occuring alpha and beta emitters like:
Th-230, Na-226, Pb-210, Pa-231, Ac-227, Ra-228 and some amounts of unrecovered U-238, U-235, U-234, Th-232 and Th-228

from the opened minerals, together with their comparatively short-lived alpha and beta decay products. Reprocessing of irradiated uranium matrix releases some unrecovered plutonium and uranium together with a number of transuranic elements. Moreover, reprocessing of irradiated natural therium matrix from fast breeder reactor also releases some Th-232, U-233, U-232 and Th-229 together with a series of short-lived alpha and beta decay products. The technology of breeding fuel is gaining importance with time.

Local or global releases of such activities can be estimated from the production figures of fissile and fertile materials. The nuclear properties of these radio-nuclides are well established for use in assessing the environmental aspects. We will not consider these radio-nuclides in the present review.

1.2 Sources of FP

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The largest group of artificial radio-nuclides, wiz FP and transuranic elements, as possible contaminant and radiation sources in the human environment, is produced by neutron induced fission as a result of exploitation of nuclear energy by man. FP disperse into the biosphere as effluents or releases principally from:

> operation of nuclear reactors, operation of fuel reprocessing plants, reactor accidents, nuclear explosions and critically excursion of fissile material mass <u>and</u>

failure of FP containments.

.3 Environmental Aspects of FPND

In the environmental considerations and quantitation f effects, one of the essential inputs is the basic nuclear ata of the FP. The relevance of FPND including neutron cross ections and yields are well established in the fields of ssessment and investigations on:

reactor and criticality accident analyses and releases,

accidents involving release of FP from containments,

nuclear explosions and releases,

build-up of FF and transuranic elements in reactors,

radioactive waste, its management and permissible offluent releases,

radiation field and shielding for protection from contained FP,

thermal output from contained FF for disposal, storage or use,

radiation field from effluent and from FP dispersed or dispersing in the environment,

energy transfer parometers from disintegrating FP into the surrounding media,

environmental distribution and ecological consequences of the presence of FP in the different matrices and biological systems,

FP chains as tracers for geochronological chemistry, biological uptake, dispersion, transport and reconcentration processes in the environment,

radiation exposure of man and routes for the appearance of FP in human system and

activation of reactor core constituents.

Methodology for utilisation of FPND are varied

and complementary non-nuclear data or informations required in the study are also extremely varied. Numerous publications

of ICRP, UNSCEAR and IAEA eminently domonstrate the methodology and its complexity in the assessment of radiation doses and resulting hazards from various sources to man. Stupendous volume of publications are available dealing with the environmental impact of the presence of FP and the resulting hasards to man and eco-systems. Hazards of FP vary with the environment and the extent and nature of its utilisation by man. To cite an example, hazards due to the presence of Sr-90 in fresh water system is much different from that due to its presence in sea water. In every environmental and regional setting including human habitation and habit, the investigator is required to discover for himself the significance of the presence of different radionuclides in their context. Global environmental consequence of radioactive releases arising from utilisation of fission process by man had so far been essentially from a

FF and neutron evaporation therefrom.

FP and unfissioned fissile materials in atmospheric explosions and

possible accumulation of Kr-85 and H-3 in the biosphere from reactors and fuel reprocessing plants.

1.4 Accuracy needed for FPND in Routine Work

It has been stated with some justification that available FPND have provided adequate informations as needed in radioactive waste management work and for purposes of dose predictions in the environment. Because of large uncertainties in:

the escape of radio-nuclides from reactor and containment systems,

fractionation during radioactive waste treatment and

setting up permissible limits for releases into the environment,

some authors are of the view that the data on FP yields need only be known to within a factor of 2 of the true data/ $\overline{2}$. It is recommended by various authors that gross calculational methods and good integral measurements under different practical situations are only needed in routine radiation protection work and in radioactive waste management. Precision data needed for routine work are half-lives, decay schemes and decay emergy spectra of FP.

1.5 Scope of the Present Review

In the present paper, we shall review the situation as to the incompleteness of the available FPND and the connected fission data of the fissile nuclides and their relevance for the environmental assessment and investigations. In nuclear reactor operation, a series of other induced radio-nuclides are formed, either in the fissioning matrix itself or in the matrix surrounding the fissioning system. Computation of the production and concentration of these induced radionuclides in the matrices require informations on neutron spectra, flux and activation cross sections for the spectra obtained in these areas. Although, integral measurement data are applicable to a particular system, discrete energy data on the above are necessary for universal application. Consideration of such nuclides in the environmental context is important, however, the paper only considers those that are fissile and have spontaneous fission half-lives low or comparable to their alpha half-lives.

and in fast reactor systems. Experimental results on the variation of $\overline{\nu}$ with incident neutron energy are available $\overline{5}$. Spectral informations of emitted neutron except for thermal neutron induced fission are not that precisely known.

2.2 Availability Status of FPND and FP yields in Binary Fission and their Use

FP of major interest in the studies of environmental aspects are those produced in binary fission. Global and local estimates of the inventory of FP from:

nuclear explosions or excursions,

nuclear reactor operations and

spontaneous fission

are to be made from:

yield data of individual FP,

scale of use of fissile materials and

production of nuclides of low spontaneous fission half-lives.

Thermal neutron induced fission is the most widely studied and sufficiently accurate data are available on mass yield, independent yield $\int 9 \int 3$ and FP activities beyond a few seconds of fission for U-233, U-235 and Pu-239. Limited data on fission neutron or fast neutron induced fission are available, though not with comparable precision. Somewhat imprecise data on prompt and delayed neutron yields for fast fission are known. While gross $\overline{\nu}$ values are well established $(1-2\%), \ \overline{\nu}(A)$ values even for thermal fission are not that precisely known. FPND of discrete energy neutron induced fission is gaining importance with the development of fast and intermediate energy reactors, nuclear explosives and excavators. Precise data on pile neutron spectrum coupled with neutron cross section data at discrete energy intervals or data obtained from

2. PERSPECTIVE FOR UTILISATION OF FPND:

2.1 Basic FFND and Use

Comprehensive compilations of nuclear properties of FP and fissile nuclides are available in a number of publications $\sqrt{3}$, 4, 5 _7 Precision data of relevance in environmental and exposure studies such as:

> Q values and nature of radioactive disintegration, decay scheme and gamma spectra, radioactive and spontaneous fission half-lives <u>and</u>

natural isotopic abundances

are available in these documents and in the nuclear data sheets published (USA) from time to time. For the current Panel Valente [5, -7] has prepared a comprehensive list of compilations on various FPND. Extensive use of such data for setting up radiation protection standards for man was made in the ICRP committee-2, UNSCEAR and in many national publications. The absorption and elimination of dispersed radio-isotopes by man and other living organisms and the concept of biological effectiveness of different types of radiations (related to the linear energy transfer rate in tissues) are used to develop the prescriptions for permissible levels of FP in the environment and in the environmental products used by man. Data on linear energy transfer rates from beta particles of a given spectrum and heavy fragments like FF in tissues are not available but FF range values are available [7, 7].

Precise experimental informations on $\overline{\nu}$ values for different fissioning nuclides - spontaneous or induced by neutrons of different energies or spectra - are of significance in the assessment of activation reactions in nuclear explosions, excumsions

integral measurements are necessary in FP computational work. Data on independent yields in induced fission by fast neutrons are still mostly to be obtained empirically or by model computation. The individual FP yield data together with the data on:

half-life,

decay-scheme,

branching ratio,

beta-decay energy and spectrum,

prompt fission gamma spectrum and neutron spectrum and

conversion electrons energy and output

are necessary for

calculation of FP inventories and shielding requirements,

evaluation of heat production,

identification and estimation of individual FP,

assessment of chemical state,

assessment of mohility of recoiling daughter species in the biosphere and in biological system and

evaluation of the hazards from accidents and explosions.

In addition to this, data on many of the short-lived precursors of FP are not available (Cf.Fig.2). Many solid FP precursors are gaseous or volatile at short times after fission. These gaseous and volatile precursors appear away from their fission domain particularly in nuclear criticality excursions or explosive reactions and as such their decay products appear as fractionated nuclei. The ratios of certain FP activities have often been used specially in fallout measurements for obtaining information on the device. It has been attempted [10] in some of the recent systematics to estimate the efficiency of an explosion from the ratio of certain FP and activation product activities to the fissile material activity, which do not undergo significant fractionation relative to one another in the fall-out. Precise yield data on FP are of importance in quantitative understanding of the fractionation and in the studies on atmospheric and environmental transport phenomenon.

Dearth of data is keenly felt for integral analysis of radiation exposure at short times after fission in nuclear accidents and explosions. It is known that total decay energy release from thermal neutron fission product gammas is almost equal to that from prompt gammas. But, because of the fast time dependence of the former, the gamma exposure at a point would be critically dependent on the time of exposure. About 25% of the energy released from FP takes place within the first 10 seconds after fission and 40% in the first 30 seconds [11, 12].

Amongst ternary FPND, fission yield data on H-3 is the most important [13] because of its biological significance and also its usefulness as a tracer in the environmental dispersion study. H-3 yield data has relevance both for nuclear explosion and for reprecessing plants handling high burn-up fuel. H-3 yield data are also important for reactors which use stainless steel cladding of fuel elements, because a significant fraction of the product tritium can disfuse through the cladding into the primary coolant.

2.4 Data on Secondary Products and Spontaneous Fission

While considering the impact of FP on the biosphere, one has also to take into account, the formation of secondary products in the fissioning matrix. For this nuclear data are required to assess the formation and yield of transuranic elements in reactors and in nuclear explosions, few of which have relatively short spontaneous fission half-lives. In the formation of transuranic elements, the cross sections

for multiple neutron capture processes are of importance and data are scarce in this area. In breeder reactors (n, 2n) reactions are also of significance in the build-up of secondary products.

With the availability of spontaneously fissioning neutron sources and also presence of these in waste streams from reprocessing plants, nuclear data on spontaneous fission are of importance in the environmental context. Experimental data on spontaneous fission half-lives are known with reasonable degree of accuracy but independent yield data on FP are imprecise.

2.5 <u>Recoil Chemistry of FP and Transuranic Elements</u>

The recoil chemistry of FP and transuranic elements in the environment has not been studied in any detail. A daughter nuclide immediately on formation can get displaced from its location by virtue of recoil and the auger shower following the process of transmutation, with or without significant recoil energy, gives rise to a very sharp local change of the states of the other chemical species or legands. The final valence state of the daughter product is greatly determined by these sharp changes and also on the chemical nature of the general environment in which transmutation has taken place. The mobility and fractionation of such nuclide in the environment, to a large extent, is then determined by the chemical state in which it finds itself as compared to the parent nuclide. The processes of chemical fractionation of FP and transuranic elements in a fluid environment are expected to influence profoundly the migration of the decay products in the environment and in the biological systems.

3. POSSIBILITIES FOR OBTAINING NEW FPND:

3.1 FP Mass Yield and Charge Dispersion

In the absence of enough experimental data on FP independent yields in fast fission, many compilers had to take recourse to certain assumptions e.g. equal charge dispersion width for all masses [9]. This assumption is known to be incorrect for the case of thermal neutron fission of U-235, but has been in use for high energy fission reactions. The charge dispersion parameters of the fragments have some times been assumed to be constant 1147 but a model study 17 shows that it does vary with fragment mass. An appropriate check for a set of charge dispersion parameters would be to calculate the total elemental yield and look for equality of yields for complementary charges [1]. Direct experimental determination of these parameters are difficult in the present state of development in experimental techniques. Information on these can come only through some empirical procedures or from computations based on theoretical models.

3.2 Beta-Gamma Activities and Half-Lives of very Short-Lived FP

In the absence of experimental data on individual FP yields and gamma spectra, in the time region of a micro-second to a few seconds after fission, recourse has to be taken to integral measurements on FP activities. Some measurements at short times after fission exist $\int 11$, 15, 16, 17.7. More detailed measurements, especially on gamma spectra for fission induced by neutrons of different energies are called for.

The gamma energy release rate calculated from the published nuclear date of individual by is found to be about two orders of magnitude lower than the experimental values, for decay times

less than 100 seconds [11,7]. This is attributed to the fact that the nuclear data was not adequately or completely available on FP of half-lives less than a few seconds. It has been estimated that the characteristics of about 100 FP, mostly short-lived having appreciable yields, remain to be identified or studied [18,7].

The data on FP are generally known only for three or four products next to the stable one in the chain. The orderdisorder model gives the limit of FP chain $\int 1_{-}^{-1} f_{-}^{-1}$ Whus, the lowest possible charge for each mass can be obtained and it gives an idea of the number of products for which data are not available. Fig. 2 (adopted from Fig.1) gives the chart of the FP decay chains. Out of a total of 671 FP tabulated for masses in the range 80 to 156 in the chart, ~ 333 remain to be identified. If one arbitrarily assumes that FP having yields less than 1% of the chain yield, are not of significance, then 243 unidentified FP would not have to be considered as important. The thick line in Fig.2 shows the boundary of FP having yields less than 1% of the chain yield.

The lower limit of composite fission product beta decay half-life has been established as 0.2 seconds [17] by integral measurements. An earlier experiment with FP gammas also gave the lower limit of composite gamma-decay half-life as 0.2 seconds [19]. One recent measurement has identified a FP (In-132) beta decay half-life as short as 0.12 seconds [20]. For each mass chain, the half-lives of its unidentified precursors have been found by extrapolation on the basis of 0.2 seconds as the limiting half-life. The gross FP beta activity calculated by including these extrapolated data is found to agree with integral measurements for decay times down to 1

milli-seconds $\int 21_{-}^{-}$. However, much smaller half-lives than 0.2 seconds for isomeric transitions exist and their identies are yet to be established.

The maximum beta energy for composite FP at 0.096 seconds of decay has been found to be 5.7 Mev / 17/. The integrated FP gamma energy release upto one second after fission was estimated to be 0.454 Mev/fission / 11/.

3.3 Ternery-Fission Yield Data

The rate of production of H-3 is around 1 atom per 1 x 10^4 to 2 x 10^4 fissions, in U-235 thermal neutron fission [22]. Spontaneous fission of Cf-252 gives around 1 atom per 4500 (± 900) fissions [23]. A detailed compilation of H-3 yields mostly for thermal neutron induced and spontaneous fission are reviewed by Cuninghame [24]. The H-3 yield from the fissioning nuclei is critically dependent on the incident neutron energy [25]. Data on termary fission yield of H-3 as a function of incident neutron energy and fissile material should be known precisely for fast systems.

3.4 Data on Spontaneous Fission of <u>Transuranic Elements</u>

Data on the formation cross-section of spontaneously fissioning nuclei as a function of neutron energy is to be known with accuracy for predicting their inventories. Integral measurements on the formation of these transuranic elements under the situations obtained in different reactor systems would be of validity in production and environmental investigations.

Nuclides like:

Cf-254, Cm-250, Cf-252, Cm-248

could be of local significance, as the spontaneous fission halflives of these are short and or comparable to their alpha decay

half-lives. Pu-240 is present in considerable quantities in high burn-up fuel. Although its spontaneous fission half-life is much longer than its alpha-decay half-life, because of its presence in high relative proportion, the neutron output from it can be of significance. Further, interest in Pu-238 is developing because of its industrial applications. It has low spontaneous fission half-life and as such its presence in environment also needs consideration.

3.5 Multiple Neutron Capture Processes and (n, 2n) Cross-Sections

The experimental difficulties are prohibitive for measurements of the formation cross-sections of many of the transuranic elements by multiple neutron capture process e.g. as in the case of nuclear detonations involving successive capture of several neutrons during the life-time of the excited state of the compound nucleus resulting from the previous capture. So, the experimentalist has to take recourse to and improve upon the integral techniques for measurements of the yields of such nuclides, formed in nuclear detonations and in high flux reactors. Further, especially for breeder system, build-up of heavy elements by (n, 2n) reactions have to be computed from theoretical estimates of the cross section $\sqrt{26}$, 27.7 in the absence of measured values.

3.6 Data on Recoil from radioactive Nuclides

Recoil energy of FP and transuranic elements and dispersion of these in the environment can be assessed when the decay schemes and yields are known with precision. Tabulated or graphical representation of recoil energy spectrum specifically of light FP and the alpha recoil of the transuranic elements would be of significant use to research chemists and biologists. For this purpose precise beta spectrum data of individual FP of interest

are necessary. There had been excellant compilations on the gamma spectra of a large number of FP but no such compilations on beta spectra are available.

3.7 Predictive Method for Primary Data on Individual FF and FP

It would be a long time, if at all, that the experimental primary data on individual FF and FP could be obtained for all the cases. Alternatively, it is likely to be more rewarding to refine the systematics and theoretical or semi-empirical models for computational purposes. The procedures that are now available $\int 28_{\odot}^{\circ}$ for calculating number of neutrons evaporated and the spectra and various other parameters need precise values on atomic masses and enutron binding energies. These are obtained from mass formulae e.g. Zeldes et al $\int 29_{\odot}^{\circ}$ and the accuracy of these in turn depends on the experimental values of atomic masses used in deriving these. Wapstra et al $\int 30_{\odot}^{\circ}$ give a table of precise atomic masses and such data on more FP are necessary to improve the accuracy of mass formulae. This envisages availability of good computer storage and retrieval facilities for FPND.

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Review Paper No. 3

THE IMPORTANCE OF FISSION PRODUCT NUCLEAR DATA IN THE

PHYSICS DESIGN OF POWER REACTOR CORES

J. G. Tyror

United Kingdom Atomic Energy Authority Atomic Energy Establishment Winfrith, Dorchester, Dorset

ABSTRACT

The paper reviews the effect of fission product poisoning on the in-core physics performance of fast and thermal reactors. The main impact is on fuel reactivity although differential effects associated with temperature and voidage are important as is the effect of Xe-135 on thermal reactor kinetics.

The sensitivity of fission product captures to yield and cross-section data is discussed. Target accuracies on fuel reactivity lifetime are set and from these the associated requirements on fission product data are deduced.

1 INTRODUCTION

For the reactor physicist concerned with nuclear core performance, the most important feature of fission products is that they capture neutrons. Such captures represent an absolute loss of efficiency in the neutron cycle and are truly parasitic. The effect of such neutron capture by fission products is generally referred to as fission product poisoning.

The most direct impact of fission product poisoning is on the fuel cycle. The presence of fission products in reactor fuel reduces the reactivity cycle length for a fuel of given enrichment or alternatively requires to be overcome by an increase in enrichment in order to maintain a given cycle length. This increase in enrichment typically adds 15-20% to thermal reactor fuel cycle costs (0.3 - 0.4 mills/KWh). In fast reactors the overall impact is smaller and is worth perhaps 0.1 mills/KWh. It is nevertheless a significant fraction of the fast reactor fuel cycle cost.

But it is not only the magnitude of the poisoning effect which is important the way in which the neutron captures take place is also significant. Thus the energy dependence of captures influences neutron spectra in the reactor and this impacts on differential effects such as reactivity coefficients. The time dependent nature of the capture processes may also be of major significance in terms of reactor dynamics and stability.

These points illustrate the fact that fission products have a significant impact on reactor design and fuel cycle costs. It follows that it is important to have a good, quantitative understanding of the various fission product effects and that in particular fission product data of appropriate accuracy are required. The present paper attempts to quantify these requirements.

2 POISONING EFFECT OF FISSION PRODUCTS

In general there are 3 potential sources of a fission product N

a direct from fission

$$S_{1} = y_{5} \int_{E} \Sigma_{f5} \not D dE + y_{9} \int \Sigma_{f9} \not D dE + \dots \qquad (1)$$

b from precursor decay

$$S_2 = N_p \lambda_p$$
 (2)

c as a result of neutron capture in another fission product

$$S_3 = N_c \int \sigma_{cs} \not B dE$$
 (3)

There are 3 potential removal routes

d as a result of neutron capture

$$R_1 = N \int \sigma_c \, \not o \, dE$$
 (4)

e as a result of radioactive decay

$$R_2 = N\lambda$$
(5)

f by physical removal or leakage diffusion from fuel

Thus, in general

$$\frac{dN}{dt} = S_1 + S_2 + S_3 - R_1 - R_2 \text{ etc}$$
(6)

In these equations the symbols have their normal meanings. In particular, y is the yield, λ the decay constant, $\sigma_{\rm C}(E)$ the microscopic capture crosssection of the fission product of atomic density N and $\beta(E)$ the neutron flux at the fission product. To simplify what is only intended to be a broad indicative discussion, we shall henceforth drop the explicit integrals over energy and write simply

$$R_1 = N \sigma_c \not 0 \text{ etc}$$
(7)

so that appropriate energy averages are to be assumed. We shall ignore capture products (is set $S_3 = 0$), assume a constant fission rate (is $\Sigma_{f} = constant$) and ignore fission product leakage in order further to simplify the treatment.

We then find that the precursor product satisfies the equation

$$\frac{dN_{p}}{dt} = y_{p} \Sigma_{p} \beta - N_{p} \lambda_{p}$$
(8)

and hence

$$N_{p} = \frac{y_{p} \Sigma_{p} \beta}{\lambda_{p}} \left(1 - e^{-\lambda_{p} t}\right)$$
(9)

$$= \frac{y_p \sum_{f} \emptyset}{\lambda_p}$$
 if the precursor decay is rapid. (10)

The equation (6) for N now becomes

$$\frac{dN}{dt} = y \Sigma_{f} \not 0 + y_{p} \Sigma_{f} \not 0 - N(\lambda + \sigma_{a} \not b).$$
(11)

Thus N =
$$\frac{(y + y_p) \Sigma_f \not a}{\lambda + \sigma_c \not a} (1 - e^{-(\lambda + \sigma_c \not a)t})$$
 (12)

and the fraction of neutrons captured in the fission product N is

$$\mathbf{F} = \frac{\sigma_c \, \mathbf{N}}{\mathbf{\Sigma}_a} \tag{13}$$

where Σ_{i} is the absorption cross-section of the reactor lattice.

We can now use the equations (12) and (13) to make some general observations about fission product captures in the period $t \leq T$ where T is the fuel dwell time in the reactor.

Saturating Fission Products

For $(\lambda + \sigma_{C} \not D)$ t >> 1, is for fission products with rapid decay (of little interest) or high cross section

$$P = (y + y_p) \cdot \frac{\sigma_o \phi}{(\lambda + \sigma_c \phi)} \langle \frac{\Sigma_f}{\Sigma_a} \rangle$$
(14)

and the neutron capture saturates at this level. The capture is seen to be directly dependent on total, cumulative yield but is less strongly dependent on capture cross-section. Indeed if

$$\sigma_{c} \not D >> \lambda \tag{15}$$

thon

$$\mathbf{F} = (\mathbf{y} + \mathbf{y}_{\mathbf{p}}) \ (\frac{\Sigma_{\mathbf{f}}}{\Sigma_{\mathbf{a}}})$$

and neutron capture is independent of cross-section.

Such high values of σ_c do occur for isotopes with resonances in the thermal region (eg Xe-135, Pm-147, Sm-149) and account for up to half the fission product captures in thermal reactors. They do not occur at energies of concern in fast reactors. The saturated fission product is thus a concept strictly relevant to thermal reactor design only.

Two saturating fission products are worthy of specific mention. The capture cross-section of X_8 -135 in the thermal energy range is enormous with a resonance at about 0.1 eV and a 2200 m/s cross-section value of about 2.7 10^6 barns. This isotope therefore saturates very rapidly. Taking values of

$$\lambda_1 = 2.1 \ 10^{-5} \ \text{secs}^{-1}$$

y+y_p = 0.065 (for U-235 fission)
 $\frac{\Sigma_f}{\Sigma_a} = 0.6$

givəs

F(10-135) -> 0.040 as \$ > 00

and at more reasonable power reactor flux levels

F(Xe-135) - 0.025.

The isotope Sm-149 also has a high capture cross-section for thermal neutrons with a 2200 m/s value of 3.8 104 barns and furthermore it is stable. Hence $\lambda = 0$ and equation (15) is always satisfied. The yield of this isotope is about 0.011 (for U-235 fission) so that the fraction of neutrons captured in this fission product soon saturates at a typical value

F(Sm-149) = 0.0075.

Non-Saturating Fission Products

Returning now to equations (12) and (13), we see that if

 $(\lambda + \sigma_{\beta} \beta)T \ll 1 \tag{16}$

is for fission products which are stable (or long lived compared with the reactor dwell time) and of small cross-section, then

$$\mathbf{F} = (\mathbf{y} + \mathbf{y}_{\mathbf{p}}) \cdot \sigma_{\mathbf{o}} \cdot (\frac{\Sigma_{\mathbf{f}}}{\Sigma_{\mathbf{a}}}) \not a \mathbf{t}$$
(17)

and the neutron capture increases linearly with time. This is the situation with fission products in fast reactors and with a large number of modest contributors in thermal reactor systems. The neutron capture is seen to be directly proportional to both yield and capture cross-section.

Total Fission Product Neutron Capture

In a real reactor situation the evaluation of fission product capture is a good deal more complex than is implied by the equations given above. Variations in fission rate, contributions from different fissioning species, and forced decay by neutron capture must be taken into account. Nevertheless the expressions given above provide a satisfactory broad picture. We note that the total fraction of neutrons which are captured in a reactor operating at steady power increases with time - roughly linearly in a fast reactor, but in a more complex manner in thermal reactors. The average fission product capture fraction in fast reactor fuel is typically 3.5% with about twice this value in fuel at the end of life. In a typical, enriched thermal power reactor, the average total fission product capture fraction is 10% and in a fuel element immediately prior to discharge, the value can be over 15%.

Target Accuracies

As stated above, the prime impact that fission product captures has on core physics design and performance is on fuel cycle length or fuel enrichment. The 1967 IAEA Panel [1] set a target of $\pm 5\%$ for the prediction of fuel reactivity lifetime in thermal reactors, and the 1971 IAEA Panel [2] suggested even tighter targets, at least for light water reactors (LWRs). Major components of such uncertainties arise from errors in modelling the reactor burn-up process and uncertainties in heavy isotope and moderator nuclear data. The contribution from uncertainties in predicting fission product captures must thus be kept small. On this basis it seems reasonable to set the target accuracy on fuel reactivity lifetime due to fission products alone at $\pm 2\%$.

In natural uranium reactors the position may be somewhat different since neutron economy is of greater importance and the reactivity lifetime is small. This situation, however, is not investigated in the present paper. Similarly, the position as regards reactors with circulating fuel [3] may be very complex but since they are relatively uncommon we will consider them no further.

In a Pu-oxide large fast reactor (FR), the presence of fission products convert what would otherwise be a small variation of reactivity with time to a significant decrease. The net effect is a loss of typically 7% reactivity at 100,000 MWd/t. An appropriate target would appear to be to be able to predict the effect to $\pm 10\%$.

THERMAL REACTORS

3 REPRESENTATION OF FISSION PRODUCTS

Several hundred different fission products arise as fuel burn-up proceeds and it is necessary to evaluate the amounts produced and the resulting neutron captures in all of them to appropriate accuracy. It is possible to identify 3 types of approach [4]:

- a A fully explicit scheme in which all products for which data are available are represented. This would involve a few hundred differential equations (eg representations in the Canadian code FISSPROD [5]).
- b A partial scheme in which those products, about 30, contributing over 90% of the total fission products are represented explicitly. The remaining, relatively unimportant products are lumped together as a single pseudo product.
- c A pseudo acheme in which all fission products, apart from the important, rapidly saturating ones, are represented in terms of 4 or 5 pseudo products (eg the Nephew [6] and Walker [7] schemes). The data for the pseudo groups are obtained by fitting to results obtained from an explicit scheme and are clearly as good as the matching involved.

In general it appears that the detail of a above is not required for physics purposes and that the value of c above is too limited, although a recent scheme involving 7 explicit groups and 3 pseudo groups (representing rapidly saturating, slowly saturating and non-saturating products) is of interest [8]. Thus the type b approach is the one frequently adopted.



Fig 1: Fission Product Chains in WIMS Library





					Fiston	Yield			Absorption	Resonance
Isotope	Identifier	Product	Product	U-235	U-238	Pu-239	Pu-24.1	Half Life	2200 m/sec (barns)	Integral 0.55 eV - 2 MeV
Kr-83	83			0.0055	6000°0	0.0029	0.0021		208	231
Mo-95	<u>8</u>			0.0655	0.056	0.050	0.048		14.1	105
To-99	66			0.0625	0.063	0.061	0.058		21.8	205
Bu-lol	101			0.050	0.063	0.059	0.061	•••••	3.6	68
Bu-103	103		103	0°029	0.062	0.056	0.063	40 days	5.0	52
Eol-48	103			0	0	0	0	•	147.0	1003
Rh-105	105		1105	0.0085	0.037	0.055	0.062	35 hrs	16000.0	24,806
Pd-105	1105			0	0	0	0		9°6	R
Pd-108	108	50T		0.000	0.0064	0.026	0.033		10.9	213
60T-3V	60T			0.0004	0.0030	0.0155	0.020		89.0	1389
	1:			0.00012	0,00095	0.0007	0.0021		20100.0	295
	15			11000.0	10000-0	0.00035	0.0005		203.0	3189
/21-1	/21			0.0025	0*0022	0.0037	0.002		0 • 2	13
TCT-OY	151			0.029	×0.0	0.038	0.030		93.0	823
C1-135	5			0,066	0.060	0.069	20.0		28.8	392
まての	\$;	5511	1	0	0	0	0	2.1 yrs	134.0	58
461-94	57		1135	0.0645	0.062	0.0715	0.078	9.2 hrs	26 70000.0	6072
C8-155	51			0	0	0	0		8.7	66
241-24				0.0575	0.0485	0.046	0.045		330.0	8
247-DR	145			0.038	01000	0.031	0.031		52.1	247
とち			2747	0.0102	0.0137	0.0095	0.0109	2.65 yrs	234.0	3028
	/***	2mm	247	6110.0	0.0153	0.0105	0.0121	2.65 yrs	234.0	3028
	2472	;		0 0	0 (0	0	1	87.0	693
		51; 1		0	0	0	0	42 days	27000.0	31615
		齐 ;		0,00			1	5.3 days	1500.0	122
		Ŗ		2010-0	norn•n	CT0.0	GTO O		5//900•0	2667
	22			0000	0	0	0		102.0	290
	101	NCT F		0100-0	0600 0	0.000	0600		12400.0	2326
201-10		201		0200.0	10000	0.000	5.5		208.0	2982
	1			GTON"	200.0	* 20*0	0.0055		390.0	1583
		CCT		0.0000					1500.0	19
				62000 0		0.0016	0.0027		14000.0	6010
107-00	<u>.</u>				/100.0	/000.0	1000-0		254000 °O	1463 201
rseado	ZNK			0.504	106+0	0.329	0.309		10.0	82
*Capture	in Pm-147 pi	roduces t	he two i	somers I	a =248 m a	hL-ad ba	8 in th	e ratio 0.	47 to 0.53.	This is

The first order, non-linear equations for the fission products are solved. together with those representing destruction and production of heavy isotopes. in a stepwise manner with occasional re-evaluation of the neutron spectra and criticality search. The WIMS code [9, 10] is typical of the approach followed. Here the basic code data library is in 69 energy groups, the 42 thermal groups below 4 eV being so spaced as to give an adequate representation of the cross-sections of the principal nuclides involved, including the resonance of Pu-240 at 1 eV. Appropriate multi-group data are available for 33 fission products. One of these is a pseudo-product which is intended to account for the 5% (approx) of fission product absorption not treated explicitly by the rest of the scheme. The product Pm-147 is, in practice, treated as 2 separate products which produce respectively the ground and metastable states of Pm-148. Table I presents a list of the WINS fission products together with their WIKS identifiers, capture and decay products, yields from fission, half lives, 2200 m/sec cross-sections and resonance integrals. The fission product chains are illustrated in Fig 1.

It has been suggested that the above scheme could be improved by the inclusion of more precursors (notably 2r-95, Nb-95, I-131, Xe-133 and Pr-143) and that a few of the existing products which are represented explicitly (eg In-115 and I-127) could be shifted to the pseudo products. The adoption of these suggestions would bring the scheme into closer alignment with that recommended by Walker [11] for the ENDFB; however, the changes would not be expected to affect the total calculated fission product absorption rate by more than about 1%.

Competition between nuclei for neutrons of thermal energy such as, for example, the effect of the presence of Pu-240 on captures in PA-147 and Sm-151 (which have resonances near to the 1 eV Pu-240 resonance, see Fig 2), is well represented in this type of approach. At higher, slowing down energies, the effect of broad flux depressions in the fuel is included but not the detailed interaction between individual resonances of the various absorbing species. As we shall see, fission product neutron capture in the slowing down region is quite significant and increases to about 30% at high burn-up. There is thus a modelling uncertainty associated with the detailed interaction effect, but it is believed to be small compared with the broad target of the previous section. Undoubtedly the major sources of uncertainty in approaches of this WIMS type are in the nuclear data.

4 INDIVIDUAL FISSION PRODUCTS

The WIMS scheme referred to above has been used to evaluate the fission product captures in typical natural uranium [12], LWR [13] and HTGCR [12] reactor lattices. The variation in the fractional capture F for the LWR lattice is shown in Fig 3 in terms of total fission products and for the major individual contributors. The differing characteristics of the saturating and non-saturating products may be clearly seen. For this case, fission products account for 12.% of all absorptions at 40,000 MWd/t. Such capture has a significant effect on the neutron spectrum both in terms of a general hardening and from local resonance effects. These spectrum effects also contribute a loss in reactivity so that at 40,000 MWd/t the presence of the fission products is found to reduce reactivity by 16%.

In Table II we compare the captures in the various reactors at a typical midirradiation point and order the important isotopes according to their relative captures. The importance of the Xe-135 contribution, especially for the low irradiation natural uranium case, is clear. It is also clear that the ordering of the isotopes is not very sensitive to reactor type and although the use of alternate data sets might give some variations in the numbers, there can be little doubt that the isotopes specifically listed are amongst the 12 most important and account for about 70% of all captures.



FIG. 3 FRACTIONAL FISSION PRODUCT CAPTURE IN A TYPICAL LIGHT WATER REACTOR LATTICE

	HTECR (60,000 HBa/t)				LWR		Naturel U	
	Low En	riched	Th/U	-235	(20,00	0 KWa/t)	(2,250	NWe/t)
	ŗ	% of Total	F	% of Tetal	F	% of Total	7	% of Total
Xe-135	0,021	20	0.021	1.7	0.021	25	C.014	38
Sm-149	300.0	8	0.007	6	0.008	10	0.005	14
Nd-143	0.009	9	0.017	14	0.007	8	0.003	8
Rh-103	800.0	8	0.007	6	0.006	7	0.002	5
Pm-147	0.006	6	0.007	6	0.005	6	0.001	3
Ie-131	0,005	5	0.007	6	0.004	5	0.001	3
Cs-133	0.005	5	0.008	6	0.004	5	0.001	3
Sm-151	0.004	4	0.004	3	0 .00 4	5	0.003	8
Sm-152	0.004	4	0.004	3	0.003	4	0.001	2
To-99	0.003	3	0.004	3	0.002	2	0.001	1
etc								
TOTAL	0.103		0.124	and the second second	0.083		0.037	

TABLE II: Fractional neutron capture by fission products in typical reactor lattices

Energy Dependence of Neutron Captures

The energy dependence of the neutron captures in fission products is of some interest in assessing data requirements. The energy variation of total fission product captures is shown in Figs 4 and 5 relative to a (1/v) dependence. The peaks around 0.1 eV due to Xe-135 and Sm-149, around 1.5 eV due to Sm-151, Pm-147 and Rh-103 and that in the 4-10 eV group due to a variety of resonances, are clear.

In Table III the fractions of captures occurring in fission products at thermal energies ($E \le 4$ eV) are given. The effect of the softer spectrum in the natural uranium case is noticeable. The energy dependence of fission product capture is also shown in Figs 6 and 7 where the reactivity contributions from captures below and above 0.625 eV are shown for LWR fuel at 20,000 MWd/t on both U and Pu enrichment [14]. Captures above 0.625 eV contribute 38% of the reactivity effect of fission products for U02-fuelled LWR and 45% for the Pu02 case. These values become 56% and 67% if we exclude Xe-135 and Sm-149.

Target Acouracies for Nuclear Data of Individual Fission Products

Having obtained an indication of the contributions of individual fission products to neutron captures, we are now in a position to assess the data accuracies implied by the lifetime target of 2% quoted earlier. A degree of judgement is necessary both on account of variations in data sensitivity with reactor type and because of the large amount of data involved - some of them correlated [15].





FIG.5 TOTAL FISSION PRODUCT CROSS-SECTION RELATIVE TO V FOR A TYPICAL H.T.G.C.R. LATTICE AT 60,000MWD/Te





	HTGCR (60,000 MNa/t)	Natural U Reactor (2,250 MWd/t)
Xe-13 5	100	100
Sm-149	100	100
Nd-143	96	99
Rh-10 3	99	99
Pm-147	27	45
Xe-131	31	47
Cs-1 33	24	43
Sm-151	96	99
Sm-152	26	44
To-99	32	53
TOTAL	73	91

TABLE III: Percentage of fission product captures in the thermal (E < 4 eV) energy range

The approach adopted here is to produce broadly consistent targets for the yield, average thermal cross-section and average resonance cross-section for individual fission product nuclides. In general terms the reactivity lifetime target is related to targets on these individual data items by an expression of the form

$$\mathbf{p}^2 = \sum_{\mathbf{i}} \mathbf{a}_{\mathbf{i}}^2 \sigma_{\mathbf{i}}^2$$
(18)

where the σ_1 represent target accuracies for yields and cross-sections and the af are weighting coefficients determined by the relative neutron captures in

the various nuclei. For a given value of p there are many combinations of of values which will satisfy equation (18). We require a set which is sensible in terms of practical achievement and we have therefore chosen solutions of the type

$$\sigma_{i} = \frac{p}{a_{i}^{2} (\Sigma a_{i})^{2}}$$
(19)

as discussed in Appendix I. This expression associates the greatest accuracy to data for these isotopes which capture most neutrons, but the square root form of the variation modifies the demands at the top of the list.

The data targets obtained from equation (19) are discussed in detail in the Appendix and are presented in Table IV. The target uncertainties on σ_c are interpreted as flux weighted averages over energy. The yields refer to total, cumulative yields and refer strictly to those from U-233, U-235 and Pu-239 thermal fission.

5 EFFECTS OF TEMPERATURE

The change of lattice reactivity with temperature is of direct concern in considerations of reactor safety, variable load operation and reactor shutdown. The change in reactivity with temperature is due to a number of factors amongst which are:
- a the effects of Doppler broadening on self-shielded resonances; and
- b changes in thormal spectrum shape due to moderator temperature changes.

For fission product nuclides, the densities are always low. Their resonances are thus unshielded so that no significant Doppler broadening effect arises. The variation with energy of fission product neutron capture, however, is significant, as we have seen. We may thus expect the fraction of fission product captures to vary as moderator temperature (and hence thermal spectrum and the balance of neutron competition) is changed.

The effect for the HTGCR lattices discussed above is shown in Table V where the variation of F for a unit temperature change is given. As expected, the effect is dominated by these nuclides with large thermal component captures. The magnitude of the overall effect is quite significant - fission product captures decreasing from 10.3% of total absorptions to 9.9% in raising the lattice temperature of the low enriched HTGCR by 100°C. The fission products thus contribute a positive component to the lattice temperature coefficient of reactivity. For this particular lattice in fact the coefficient is $-6.4 \ 10^{-5/\circ}C$ and in the absence of fission products, the value would be $-10.0 \ 10^{-5/\circ}C$. The target accuracy on temperature coefficient is about $\pm 0.5 \ 10^{-5/\circ}C$ and in the present, probably extreme example, this is consistent with knowing the fission product component to $\pm 10\%$.

The Xe-135 contribution is the most significant, accounting for about half this effect. We thus take the target accuracy for this Xe-135 component itself to be about 10%. The yield target of Table IV is clearly well within the figure but the real implication of the temperature effect is the requirement on the shape of the cross-section curve. This is impossible to quantify simply in

	Yield y	Thermal Cross-section $\sigma_{\rm c}$	Resonance Cross-section RI
Xe-135	5	8	(100)
Sm-149	8	(20)	
Nd-143	6	6	30
Rh-103	6 ^a	6	(50)
Pm-147	7	15	8
Xe-131	8	15	10
Cs~133	8	15	10
Sm-151	8 ^b	8	40
Sm-15 2	9 ^b	20	10
Tc-99	11	20	15
etc			

Notes: a Yield is effectively that of Ru-103 b Effective yield including decay and neutron capture yield

TABLE IV: Target accuracies (in percentage of value) for fission product data in thermal reactors view of the complex nature of the temperature effect, but clearly the position of the Xe-135 resonance and the energy variation of the associated capture cross-section is required to an accuracy consistent with the above 10% figure. It is probable that the 8% target accuracy of Table IV, interpreted now as applying at all energies in the range 0.05 - 0.2 eV is sufficient. From similar considerations we may also derive a need for information on the shape of the $\sigma(E)$ variation for other isotopes. For Sm-149 we assess the requirement at $\pm 20\%$ in relative values over the energy range 0.06 - 20 eV.

	Low Enriched		U/Th		
	$\frac{\Delta F}{\Delta T} \ge 10^{-5}/°c$	% of Total	$\frac{\Delta F}{\Delta T} \times 10^{-5}/^{\circ}C$	% of Total	
Xe-135	1.90	46	1.20	71	
Sm-149	0.65	16	0.34	20	
Nd-143	0.38	9	0.10		
Rh-103	-0.07		-0.21	-12	
Pm-147	0.03		0		
Xe-131	0.07		0.02		
Cs-133	0.05		0.03		
Sm-151	0.39	10	0.29	17	
Sm-152	0,03		0		
Tc-99	0.03		0		
TOTAL	4.1		1.7		

TABLE V: Change in fractional fission product capture with temperature for HTGCR lattices at operating temperature and at an irradiation of 60,000 MWd/t

6 KINETIC AND STABILITY EFFECTS

The time constants associated with the production and removal of Xe-135 are measured in hours so that variation in the density of this poison may have an impact on relatively short term reactor kinetics. The presence of this poison and its delayed production have a destabilising effect on reactor power since, for example, any increase in power leads to burn-out of Xe, a gain of reactivity and further power rise before the effect of increased precursor I-135 production is felt. The reactor design implications of (uncontrolled) unstable behaviour are significant and extend into the control and instrumentation areas.

The question of xenon instability has been much discussed in the literature. The contribution made by Xe-135 to the reactor stability criterion is approx (see [16])

$$Q \propto \frac{(\mathbf{y}_{I} + \mathbf{y}_{x}) \left[\frac{\sigma_{x}}{\lambda_{x} + \sigma_{x}} - \frac{\mathbf{y}_{x}}{\mathbf{y}_{I} + \mathbf{y}_{x}} \right] \phi_{\sigma_{x}}}{\lambda_{I} + \lambda_{x} + \rho_{\sigma_{x}}}$$

(20)

where suffices x and I refer to Xe-135 and I-135 respectively. In practice, of course, sophisticated computer codes are used to evaluate stability thresholds and associated transients. The data requirements of such models may, however, be assumed to be such that the parameter Q (which at high flux depends only on y_I) should be capable of evaluation to about $\pm 10\%$. This is well within the targets set previously on fuel reactivity criteria.

Variations in power level lead to variations in Xe-135 density and hence in the reactivity held by this poison, and such variations need to be overcome by an appropriately designed control system. The maximum effects are those associated with changes down to zero power level. In these circumstances the reactivity held by Xe-135 rises to a maximum in about 10 hours and then decays away to zero on the Xe-135 decay period. In fact we find

$$\mathbf{F}(\mathbf{Xe-135,t}) = \left(\frac{\Sigma_{f}}{\Sigma_{a}}\right) \left[\frac{y \phi_{o} \sigma_{c}}{(\lambda_{I} - \lambda_{x})} \left(e^{-\lambda_{x} t} - e^{-\lambda_{I} t}\right) + \frac{y \phi_{o} \sigma_{c}}{(\lambda_{x} + \phi_{o} \sigma_{c})} e^{-\lambda_{x} t}\right]$$
(21)

where $y = y_{I}+y_{X}$ is the cumulative yield of Xe-135 and β_{0} is the flux level prior to shutdown. Thus, as well as depending directly on yield and $(\lambda_{I}-\lambda_{X})$, the value of F is also strongly dependent on σ_{c} , even at high values of β_{0} . Typically F(t) may rise to twice its normal value so that if it is desired to start up the reactor at this point in time, a control system capable of compansating for an additional Xe-135 reactivity effect of 2-3% is required. Design of such a system requires a knowledge of F(t) to about $\pm 10\%$. Given the yield and cross-section targets of Table IV, this requirement implies a need for values of λ_{T} and λ_{-} to a target accuracy of $\pm 5\%$.

Other fission products have a much smaller impact on neutron kinetics. The Sm-149 level increases after shutdown as Pm-147 decays, is

$$\mathbf{F(t)} = \mathbf{F(o)} \left(1 + \frac{\sigma \not o}{\lambda_p} \left(1 - e^{-\lambda_p t}\right)\right)$$

but even after a long shutdown, the increase in F is only about 20%. The effects need to be taken into account but they make no new demands on nuclear data.

The importance of delayed neutrons is discussed in Appendix II.

FAST REACTORS

7 REPRESENTATION OF FISSION PRODUCTS

Commercial Fast Reactors are being designed using fuel which is required to go to high burn-up - typically 100,000 MWd/t. This burn-up implies the fissioning of 10% of heavy atoms and hence the fission products produced will total 20% an amount greater than the amount of fissile material present. The quantity of fission products present in a fast reactor will thus be significant. However, the cross-sections of these fission products at neutron energies of interest are low. Typically the cross-section of an average fission product pair is only about 10% of the Pu-239 fission cross-section at 100 KeV. The amount of fission product capture and its effect on reactivity is thus relatively small.

In Section 3 on Thermal Reactors we noted that individual representation of several of the most important fission products was required in view of their saturating and resonance capture characteristics. This is not the situation with Fast Reactors. Here, the low cross-section for fission products implies non-saturation, and there are no dominating resonance effects. Furthermore, the present high level of data uncertainties associated with individual fission products does not readily justify a detailed, individual representation. In these circumstances, it is fairly common practice to use a single pseudo fission product or a lumped cross-section model. In this model an average microscopic cross-section over all products is used to characterise a single pseudo product and this cross-section is assumed to be independent of burn-up. The crosssection is represented in a multi-energy group form in a manner consistent with the overall scheme of data and neutron spectrum representation.

The lumped cross-section model implies no variation with time in the relative concentrations of individual fission products. In practice changes will occur due to B-decay, neutron capture and perhaps even fission product gas leakage. The most important β -decay chains appear to be those involving atomic numbers A study of the overall effect suggests that an increase of 103. 131 and 133. about 5% in the lumped cross-section over a burn-up of 300 days is strictly The position on neutron capture is rather similar. required on this account. Those fission products with the highest cross-section do experience (n, γ) conversion and the burn-up of these products at 100,000 MWd/t can reach 50% However, the new product is often itself a direct fission product [17]. and in any event there is no major variation in cross-section from isotope to isotope.

Comparisons between results obtained from the lumped cross-section model and detailed individual product representations [18] suggests that the former is uncertain up to about 5% in total fission product captures and this needs to be borne in mind when assessing data requirements. It is clear, however, that the detailed representation model is available for use as and when the needs arise and for this model the major uncertainty is in the fission product data.

8 INDIVIDUAL FISSION PRODUCTS

With Thermal Reactors we saw that the requirements for fission product data were largely set by their contribution to fuel reactivity changes with burn-up. The same is true for Fast Reactors. In the absence of fission products, the reactivity of a fast reactor changes little with burn-up due to the Pu breeding capability. The effect of fission product poisoning leads to a marked decrease as illustrated in Fig 8 [19] for a 600 MW(E) fast reactor using PuO₂ fuel of LWR isotopic discharge composition. The effect of the fission products at 100,000 MWd/t is to reduce reactivity by 7% and the effect in a batch fuelled reactor is about 5% in reactivity. As with thermal reactors, a knowledge of this value is important in fuel cycle and control design features. We shall take as our target the need to evaluate the effect of fission products in a reactor to within 0.5% of reactivity, is to $\pm 10\%$ precision.

This target accuracy of 10% in fission product captures is looser than the assessed accuracy of the lumped cross-section model. Thus continued use of this model implies the need for fission product data which contributes no more than a further 8% uncertainty to the magnitude of the total capture fraction F. This implies an accuracy of 8% for the lumped cross-section and this is not inconsistent with Greebler's 1975 goal [24]. On the other hand, use of models in which the most important individual decay and capture chains are explicitly represented would lead to a small relaxation in the data requirements.

The total fraction of fission product captures increases roughly linearly with irradiation. At any particular irradiation the relative contribution from individual isotopes is approximately determined by the product of yield and cross-section averaged over the neutron spectrum and more precisely determined by explicit representation models. In Table VI [20] the fission products are listed in decreasing order of contribution for a large 1,000 MW(E) PuO₂ reactor using Cook's cross-section file [21] and Meek and Rider yields

Fission Product Group	No of Elements	Percent of Total Fission Product Captures	Fission Products in Order of Decreasing Magnitude of Isotope Capture
I	13	60	Ru-101, Rh-103, Tc-99, Cs-133, Pd-105, Ru-102, Pd-107, Xe-131, Nd-143, Pm-147, Sm-149, Sm-151, No-97
II	13	22	No-95, Ca-135, Nd-145, No-98, Ag-109, Ru-104, Eu-153, Pr-141, Xe-132, No-100, Sm-152, Bu-154, Zr-93
ш	13	8	Sm-147, Nd-148, Ru-103, Pd-106, La-139, Nd-144, Bu-155, Nd-146, Xe-134, I-127, Pd-104, I-129, Pd-108
IV	13	4.5	Ru-100, Zr-96, Cs-134, Ce-142, Sm-150, Zr-91, Ru-106, Kr-83, Nd-150, Ce-144, Ce-140, Cs-137, Sm-148
V	13	1.9	Cd-111, Rb-85, Sm-154, Gd-156, Gd-157, In-115, Gd-155, Br-81, Sb-125, Nb-95, Pr-143, Zr-94, Zr-95
TOTAL	65	%•4	
VI	113	3.6	
TOTAL	178	100	

TABLE VI: Grouping of Fission Products according to their relative neutron capture contribution in a typical Fast Reactor

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FIG.8 EFFECT OF FISSION PRODUCTS ON REACTIVITY CHANGE IN A PuO2 FAST REACTOR LATTICE.

[22]. Similar information is given in Table VII [17] where fission products are ordered for their capture contributions in the LMFBR benchmark (ZPR-6, Assembly 7) spectrum using ENDF/B III cross-sections and the yield data of Flynn and Glendenin [23]. The magnitudes of the per-atom worths given in Table VII may be compared with values of about 80 for Pu-239, 5 for U-238 and 0.3 for Na.

The ordering of the individual isotopes is broadly similar in the two cases and whilst different data sets and spectra will produce somewhat different lists, we shall base our evaluation of the data targets for individual isotopes on the values quoted in Table VII.

Mass No	Bloment	% of Captures	Per-Atom Worth (Arb Units)
101	Bu	8.2	13.0
101	Dh	7.8	
105	Pa	7.2	74.0
133	Ca.	7.0	11.8
99	To	5.9	-
102	Ru	5.0	6.6
31.7	Fm/Sm	1.3	28.0
151	Sm/Rn	1.3	65.1
107	Pa	4.1	11.9
131	Xo	3.7	10.3
7).2	18 <i>a</i>	27	30.2
07	lia. Vo	J•1 3.6	6.7
71		3.5	5.7
110	8.	2.5	36.2
05	Mo	2.9	6.5
, ,,	27	J •2	
104	Ru	2.8	3.0
100	Mo	2.5	2.3
98	No	2.4	3.0
106	Ru/Pd	2.4	-
141	Pr	2.3	4.4
145	Na	2.3	9.3
139	La	1.9	2.2
153	Eu	1.8	52.2
109	Ag	1.7	11.7
137	Cs	1.7	1.1
152	Sm	1,1	24.7
155	Eu/Gd	1.0	63.3
157	Gđ	0.8	
L	Lauran	i	Le maile ser anne and anne anne anne anne anne anne

TABLE VII: Fission product isotopes ordered by yield-reaction rate product in a Fast Reactor

Data Target Accuracies

We noted in Section 4 that a considerable quantity of data in the form of differential cross-sections and yields is involved in evaluating the total fission product poisoning effect. We shall focus simply on the yield and average capture cross-section for each nuclide. The relationship between the uncertainty in the total poisoning, F, and uncertainties in the data associated with the ith fission product is given by

$$\left(\frac{\mathrm{d}\mathbf{F}}{\mathrm{P}}\right)^{2} = \left(\mathbf{F}_{i}\right)^{2} \left(\left(\frac{\mathrm{d}\sigma}{\sigma}\right)^{2} + \left(\frac{\mathrm{d}y}{\mathrm{y}}\right)^{2}\right)_{i}$$
(22)

This is of the same form as equation (18) and, as with the discussion of thermal reactor requirements, we select the form of solution given by equation (19) using as an overall target

$$\frac{\mathrm{dF}}{\mathrm{F}} = 0.08. \tag{23}$$

The data accuracies derived using equations (19), (22) and (23) are given in Table VIII. It will be seen that for no isotopes are the yield (from Pu-239 fission) and cross-section required to better than 20%. It should be emphasised however that this distribution of target precisions between the isotopes of interest is but a plausible one and many others are possible. It assumes that it is preferable to aim at modest (20-30%) precision for several isotopes rather than high (approx 10%) precision for a few. The views of measurers and evaluators on this point are clearly relevant.

It is interesting to note that of the two 'top ten' lists of Tables IV and VIII, no less than 6 isotopes (Tc-99, Rh-103, Xe-131, Cs-133, Pm-147 and Sm-151) are common to both. The yield accuracy requirements are, however, significantly tighter for the requirements of thermal reactors. The cross-section values and their targets in Table VIII refer strictly to per-atom reaction rates in a fast reactor neutron spectrum and are thus to be interpreted as appropriately energy averaged values.

9 THE EFFECTS OF FISSION PRODUCTS IN FAST REACTORS

In the previous section we discussed the effect of fission products on reactivity in a fast reactor and set out data targets on this basis. The presence of fission products, however, has other effects which stem from the fact that the poisoning effect is energy dependent. The fission product captures thus influence spectrum slightly and, more important, any change of spectrum leads to a change in fission product capture. This is particularly significant in the case of the sodium voiding effect. The presence of the Na coolant in a fast reactor has a major influence on the general shape of the neutron spectrum due to its slowing down power. If the Na is lost, then the spectrum is hardened considerably and the fractional capture in fission products is reduced since most capture takes place at low energies (see Fig 9). The loss of Na thus leads to an increase in reactivity, a considerable proportion of which is due to fission product effects, and a knowledge of the magnitude is important in control and safety arguments.

Typical values of a number of parameters for a typical 600 MW(E) Pu02 reactor at mid-lifetime irradiation of 50,000 MWd/t are given in Table IX [19]. The effect of fission products on the reactivity coefficients associated with Na voiding and temperature change are seen to be significant but for other parameters the impact is small. In particular, there is no analogy in fast reactors with the kinetic effect of Xe-135 in thermal reactors.

	Typical V	Jalues	Target	
Isotope	Chain Yield [24] %	Cross-section Arbitrary Units	Precision %	
Ru-101	6.9	40	20	
Rh-103	5.8	44	20	
Pa-105	5.7	42	20	
Cs-135	6.9	34	20	
To-99	5.9	33	20	
Ru-102	7.0	24	25	
Pm-14 7	2.0	69	25	
Sm-1 51	0.8	167	25	
Pd-107	3.7	37	25	
Xe-131	4.1	30	30	
Na-143	4.3	22	30	
Mo-97	5.5	22	30	
Cs-135	7.5	16	30	
Sm-149	1.4	85	30	
№0 95	4.8	22	30	
Ru-104	6.8	14	30	
Mo-100	6.8	12	35	
Mo~ 98	5.8	14	35	
Ru-106	4.7	11	35	
Pu-141	5.1	15	35	
Na-145	3.0	25	35	
La-139	6.0	10	40	
Eu-1 53	0.5	126	40	
Ag-109	1.6	34	40	
Cs-137	6.6	8	40	
Sm-152	0.7	52	55	
Eu-155	0.2	161	55	

TABLE VIII: Typical values and target accuracies in percentage of value for fission product data in fast reactors derived assuming:

- a total fission product captures required to ±8%;
- b allocation of targets between isotopes inversely as the square root of isotope capture



The results given in Table IX suggest that the reactivity held by fission products in an equilibrium, mid-lifetime core reduces from 3.5% to 2.8% as a result of complete Na voiding. We require to evaluate this change to about 0.2% reactivity, is to obtain the change in fission product captures to about 30%. The effect is determined by the energy dependence of fission product captures at low energy in the shape of the cross-sections in the energy range 0.1 to 100 KeV. The target accuracies of Table VIII appear adequate for this purpose. Values of Doppler temperature coefficient are less sensitive to fission product capture and whilst the effects are important, the fission product data requirements are less onerous than those of Table VIII.

	With Fission Products	Without Fission Products
Na voiding (Reactivity %)	1.3	0.6
Doppler Temperature Coefficient $(\frac{TdK}{dT})$	-0.013	-0.016
Internal Breeding Ratio	1.011	1.010
Effective Delayed Neutron Fraction	0.00381	0.00383
Mean Neutron Lifetime (10^{-7} secs)	4.0	4.2

TABLE IX: Effect of the presence of fission products on fast reactor parameters

10 SUMMARY AND CONCLUSIONS

The in-core physics requirements for fission product data are set by reactivity considerations in both fast and thermal reactors. These requirements have been quantified against plausible targets and the results presented in Tables IV and VIII.

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APPENDIX I

TARGET UNCERTAINTY ANALYSIS

For thermal reactors we consider a low enriched reactor, such as an LWR, in which reactivity falls linearly with burn-up, ie

$$\mathbf{k} = \mathbf{k}_{o} - \mathbf{S} - \mathbf{at} - \mathbf{a}_{p}\mathbf{t}$$

where

ε.

k = initial lattice reactivity

S = reactivity worth of rapidly saturating fission products

 α = rate of loss of reactivity due to fuel depletion, etc

 $\alpha_{\rm p}$ = rate of loss of reactivity due to non-saturated fission products.

The reactivity lifetime T is given by

$$k_1 = k_0 - S - (\alpha + \alpha_p)T$$

where k, is the lattice reactivity at end of cycle, ie.

$$T = \frac{k_0 - S - k_1}{\alpha + \alpha_p} . \tag{A.1}$$

Variations in fission product effects thus produce a variation in reactivity lifetime given by

$$\frac{dT}{T} = -\left(\frac{S}{k_0 - S - k_1}\right)\frac{dS}{S} - \left(\frac{\alpha_p}{\alpha + \alpha_p}\right)\frac{d\alpha_p}{\alpha_p} \quad . \tag{A.2}$$

Typical values for an LWR might be

$$k_{o} = 1.25$$

 $S = 0.03$
 $\alpha = 0.055 \text{ per } 10,000 \text{ MWd/t}$
 $\alpha_{p} = 0.035 \text{ per } 10,000 \text{ MWd/t}$
 $k_{1} = 0.95$

ie saturated poisons account for about 10% of the reactivity loss during burn-up and the build-up of fission products accounts for about 40% of the slope of the reactivity variation with burn-up. Thus

$$\frac{dT}{T} = 0.12 \frac{dS}{S} + 0.4 \frac{d\alpha_p}{\alpha_p} . \qquad (A.3)$$

The saturated poisons of interest are Xe-135 and Sm-149. Thus

$$S = Ay_{x} \left(\frac{\beta \sigma_{x}}{\lambda_{x} + \beta \sigma_{x}} \right) + By_{S}$$
 (A.4)

$$\frac{dS}{S} = \frac{3}{4} \left(\frac{dy}{y} \right)_{Xe-135} + \frac{1}{4} \left(\frac{d\sigma}{\sigma} \right)_{Xe-135} + \frac{1}{4} \left(\frac{dy}{y} \right)_{Sm-149}$$
(A.5)

For the non-saturating poisons

$$a_{p} = \sum_{i} y_{i} (\sigma_{i} \not \theta_{i} + (RI)_{i} \not \theta_{2})$$
(A.6)

where we have separately identified thermal and resonance fission product capture, and summation is over all relevant fission products. Thus

$$\frac{\alpha_p}{\alpha_p} = \sum_i \left(\frac{dy}{y}\right)_i f_i + \Sigma \left(\frac{d\sigma}{\sigma}\right)_i f_{i1} + \Sigma \left(\frac{dRI}{RI}\right)_i f_{i2}$$
(A.7)

where f_{i1} , f_{i2} ($f_{i1} + f_{i2} = f_i$) are the fractional contributions to α_p from captures in the ith fission product at thermal and resonance energies respectively.

We may now use equations (A.3), (A.5) and (A.7) to relate uncertainties in fission product data to the target lifetime uncertainty. Assuming no correlation we have

$$\left(\frac{\mathrm{d}\mathbf{T}}{\mathrm{T}}\right)^{2} = \left[0.12 \times \frac{3}{4} \left(\frac{\mathrm{d}\mathbf{y}}{\mathrm{y}}\right)_{\mathrm{Xe-135}}\right]^{2} + \left[0.12 \times \frac{1}{4} \left(\frac{\mathrm{d}\sigma}{\sigma}\right)_{\mathrm{Xe-135}}\right]^{2} + \cdots$$

$$\cdots + \left[0.4 \mathrm{f}_{\mathrm{i}} \left(\frac{\mathrm{d}\mathbf{y}}{\mathrm{y}}\right)_{\mathrm{i}}\right]^{2} + \cdots$$

$$(A.8)$$

This equation is of the general form

$$\mathbf{p}^2 = \sum_{i} \mathbf{a}_{i}^2 \sigma_{i}^2 \tag{A.9}$$

For a given value of p, there are many combinations of σ_i values which will satisfy equation (A.9). One such solution, in which all terms on the RHS of equation (A.9) are equal (is all uncertainty components contribute equal variance) is given by

$$\sigma_{i} = \frac{p}{a_{i} \sqrt{N}}$$
(A.10)

where N is the number of terms. There are typically of the order of 100 data items in an approach of the WIMS type so that

$$\sigma_{i} = \frac{p}{10 a_{i}}$$
(A.11)

This is a perfectly legitimate and commonly adopted solution which has the desirable property of associating the greatest precision with those isotopes which capture most neutrons. The precision required of these isotopes may, however, be unduly severe and thus very expensive to obtain. Indeed if we assume that the cost of achieving a precision σ_1 is inversely proportional to σ_1 , then the total cost of achieving our data targets is

$$c = A \cdot \sum_{i=\sigma_{i}^{2}}^{1}$$
(A.12)

and we should search for solutions of equation (A.9) which minimise the value of c. Following normal minimisation procedures we have

$$\frac{\partial}{\partial \sigma_{i}} \begin{bmatrix} A \sum_{i} \frac{1}{\sigma_{i}^{2}} - p^{2} + \sum_{i} a_{i}^{2} \sigma_{i}^{2} \\ i \sigma_{i} \end{bmatrix} = 0$$
 (A.13)

so that

$$\sigma_{i} = \frac{p}{a_{i}^{\frac{1}{2}} (\Sigma a_{i})^{\frac{1}{2}}}$$
(A.14)

This is an alternative solution to that given by equation (A.11) and again associates greatest precision with the most important isotopes although the weighting is less severe. The expression given by equation (A.14) is the one used throughout this paper to derive values of σ_1 . For thermal reactors

$$\mathbf{p} = \frac{\mathrm{d}\mathbf{T}}{\mathrm{T}} = 0.02$$

and the f_i values of equation (A.8) are obtained from the LWR results discussed in the paper. The associated values of σ_i obtained from equation (A.14) are given in Table IV where the yield referred to is the net, cumulative yield.

APPENDIX II

DELAYED NEUTRONS

Delayed neutrons arise during the decay process of certain fission products and play a well recognised and important role in reactor kinetics [16]. In power reactor kinetic and safety studies many physical processes need to be represented - initiating mechanisms, control responses, temperature changes, reactivity feedback - as well as the delayed neutron effect. Knowledge of these processes is often not good and the modelling of the event may also be associated with considerable uncertainty. Furthermore, detailed representation of the delayed neutron phenomenon is only required for very rapid transients which are extremely unlikely to occur. In all these circumstances, knowledge of the delayed neutron effect is not required to high precision and it is judged that existing data are of adequate accuracy.

The more onerous requirement for delayed neutron data precision occurs in reactor physics experiments where period measurements are made and require to be interproted in terms of reactivity. Such experiments are performed in zero or low power reactors where it is required to evaluate, for design purposes, the reactivity changes associated with various materials, reactor states, etc. Review Paper No 14 reviews experiments of this type specifically aimed at measuring the effect of fission product poisons. The interpretation of flux period in terms of absolute reactivity for direct comparison with prediction depends directly on a knowledge of the total delayed neutron fraction. For the cleanest situations, the overall precision associated with measurements of this type may be limited by uncertainties in β . Thus the interpretation of control reactivity worth in the fast ZEBRA reactor has recently been significantly improved by adopting newly evaluated &-values. For those concerned with absolute reactivity measurements, the precision target on the thermal B-values for U-235, U-233 and Pu-239 and the fast 8-values for U-235, U-238 and Pu-239 is set at +5% and thus is consistent with requests submitted to the RENDA list.

IMPORTANCE OF FISSION PRODUCT NUCLEAR DATA

FOR ENGINEERING DESIGN AND OPERATION OF REACTORS

C. DEVILLERS

Service d'Etudes de Réacteurs et de Mathématiques Appliquées

CENTRE D'ETUDES NUCLEAIRES DE SACLAY

Abstract : Fission product nuclear data and accuracy requirements are reviewed in the following fields of reactor design and operation :

- fission product release and contamination of reactor components
- fuel failure detection and Location
- fission product heating after shutdown

Several power reactor types are considered and their particular features with respect to fission products emphasized.

1 - INTRODUCTION -

This paper is devoted to review the requirements for fission product nuclear data in the three following fields of engineering design and operation of reactors :

- 1. fission product release and contamination of reactor components
- 2. fuel failure detection
- 3. fission product heating after shutdown

The two first questions are very similar in that respect that fission products are important not only in function of their nuclear properties, fission yields, half-lives, γ -ray emission, but also in function of other factors, which may be different from one reactor type to another, and which govern their transport from birthplace to the appropriate parts of the reactor where their activity is to be predicted or measured. We will therefore rely upon the experience of reactor designers when deriving sets of important fission products without extensive justification.

For the question of fission product heating after shutdown on the contrary, given cooling time regions where the heating function has to be known, one can in principle derive lists of important fission products from complete fission product data files and relevant computer codes as some examples will illustrate below.

In addition to the relative importances of the different fission products which may be helpful to set up priorities, we have tried to situate their absolute importance compared to other competing radiation sources in order to justify accuracy requirements.

Before speaking about the adequacy of present fission product nuclear data status of knowledge it must be realized that the first problem engineers have been faced with was to collect and to select nuclear data, starting from the existing compilations, and that there has been an efflorescence of home-made libraries. Sometimes engineers could afford being helped by nuclear physicists as we did, sometimes not. It is therefore difficult to appreciate the status of knowledge of fission product nuclear data through particulate libraries.

However we will give some examples of sensitivity studies based on our own codes [1] [2] and associate library [3] in order to help further discussions.

2 - FISSION PRODUCT RELEASE AND CONTAMINATION OF REACTOR COMPONENTS

The up-to-date question of reactor contamination raises as how to predict and to control the distribution of fission products escaping a certain fraction of the fuel volume and how to design the plant in order to keep the man-power pool within radiation exposure limits. To-days typical

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design fuel failure rates range from 10^{-4} for HTGR, 10^{-3} for LMFBR to 10^{-2} for PWR and BWR ; this results in a continuous leakage of fission products into the coolant circuit. In addition, fission products may diffuse through the cladding like tritium in stainless steel. Fission products released from the fuel spraid in the circuit where they accumulate either in the coolant or on the circuit wall. The circuit activity is usually limited by cleaning up a fraction or even the totality of the coolant flow. Other operations such as renewing water for boron control in PWR, removing radiolyse gases for example, transfer radionuclides outside the circuit.

There they create another problem because they must be retrieved and stored before being released outside the plant ; these environmental aspects are treated in paper n°2.

From the point of view of contamination problems inside the reactor building, fission products can be classified in two groups :

1. The first group will contain short-lived "volatile" isotopes which can leave rapidly failed fuel rods (or particles); these are noble gases with halflives greater than 1 minute and iodines 131 I, 133 I the latter giving rate to 133 Xe.

They represent roughly 10 % of the total fuel activity during operation and they will be responsible for radiation level in case of leakage of the circuit in accessible areas of the plant (in particular in case of accident). The case of the 10years half-life 85 Kr can be regarded more as an environmental problem than a contamination's one.

2. The second group contains isotopes with half-lives greater than 10 days, high cumulative fission yields; they or their daughter nuclei are hard gammaray emitters, some are very long-lived beta emitters.

Some of them have low yield values but can be produced by neutron capture on stable or long lived nuclides :

109 Ag (n,
$$\gamma$$
) 110 m Ag
133 Cs (n, γ) 134 Cs
135 Cs (n, γ) 136 Cs
147 Pm (n, γ) 148 m Pm
148 Pm

Those of the second group isotopes which will succeed in leaving failed fuel and plating out before being submitted to cleaning up will be responsible for contamination of reactor components ; 131 I which has been already mentioned as belonging to the first group can also be regarded as an important nuclide for reactor component contamination.

In table I are reported for illustration, results of measurements of important fission product activity in the circuit of the CNA 250 MWe PWR during constant power operation [4] [5].

Increasing the accuracy of fission products nuclear data cannot be justified to-day from the point of view of a better knowledge of the consequences of hypothetical accidents because :

- 1. the quantities of fission products released in case of accident and the way they distribute are by far more uncertain than their nuclear data.
- even in case of pessimistic assumptions on the release, the risks that accident represent for plant personal are less than the hazards they are exposed to during normal reactor operation.

A large fraction of radiation exposure limit is delivered to manpower pools in light water reactors during refuelling or maintenance outages; e.g. last 1973 outage of CNA costed 150 man.rem " [7]. When the circuit is opened on the storage pool for refueling, within a few days after shutdown, main radiation and body contamination sources come from short-lived nuclides of Table I, especially 131 I and 133 Xe. A thorough investigation of fission product release rates after shutdown is being done [5] [8] which might justify predictions of fuel inventory to within \pm 50 % accuracy for important isotopes. Deposition of long-lived nuclides on pipes, valves pumps and heat-exchangers is now responsible for 80 % of the radiation exposure for inspection work at the CNA although the actual fuel failure rate is far from the 10⁻² design value.

In sodium cooled fast breeder reactors a fraction of the volatile isotopes migrates towards the argon circuit, over the sodium pool ; 133 Xe, 135 Xe will govern the biological hazard in case of argon leakage in accessible areas whereas 87 Kr, 88 Kr will impose the shielding requirements [9] around the argon circuit (2000 tons concrete in Phenix).

×

the annual radiation exposure limit is 5 rem

For fuel failure rates of 10^{-3} it is estimated that long-lived fission product deposition will contribute to dose rate with about the same magnitude as corrosion products such as 54 Mn, 58 Co, 60 Co [6] [10].

TABLE 1

RELATIVE ACTIVITIES OF FISSION PRODUCTS IN THE COOLANT OF THE CNA^{*}250 MWe PWR

ISOTOPE	HALF-L1FE	RELATIVE ACTIVITY
······································	5	:
: 85 m Kr	: : 4.4 h	: 3 :
88 Kr	2.8 h	• • • •
: 131 I	: 8.05 d	0.7
: 132 I	2.3 h	: 1.4 :
: 133 I	21 h	2.5
: 134 I	52.8 m	: 1.1 :
: 135 I	6.7 h	0.8
133 Xe	5.27 d	67
: 135 Xe	9.2 h	: 17 :
: 138 Cs	32 m	3
•	•	;
: 95 Zr	65 d	23
103 Ru	39.6 d	17
134 Cs	2.19 y	28
: 137 Cs	30 y	32

X

CNA : Centrale Nucléaire des Ardennes

In helium cooled high temperature reactors one can expect that fission products will be dominant in contamination problems due to the absence of metal structure in the core, important isotopes being 90 Sr, 131 I, 134 Cs, 137 Cs.

Summarizing the very similar lists of fission products which are recognized as important, by contributors to this paper, gives the following :

1. volatile isotopes of half-life ranging from 1 minute to 10 days : noble gases : 85 mKr, 87 Kr, 88 Kr, 89 Kr 133 Xe, 135 mXe, 135 Xe, 137 Xe

iodines : 131 I, 133 I

2. isotopes of half-life greater than 10 days :

noble gases : 85 Kr

solid isotopes : 90 Sr, 95 Zr, 103 Ru, 106 Ru, 110 m Ag, 125 Sb, 129 mTe, 134 Cs, 136 Cs, 137 Cs, 140 Ba

To this list must be added three types of related nuclides :

2.a : stable or long-lived precursors by neutron capture :

2.b : short-lived beta decay gaseous precursors which may govern the transport of solid isotopes :

> 90 Kr ______ 90 Sr 137 Xe ______ 137 Cs 140 Xe ______ (140 Cs)_____140 Ba



The types of nuclear data which are requested for each of these isotope lists are indicated in table II.

TABLE II

TYPES OF NUCLEAR DATA NEEDED FOR EACH ISOTOPE LIST

:	List	Cumulative Vield	Half-life	Neutron absorption cross-section	y-ray spectrum	Decay branching ratio from parent	Neutron capture cross-section	Capture branching ratio from parent
2 2 2 2 2 2 2 2 2 2 2 2 2 2 2 2 2 2 2	1 1-a	+ + : +	; ; ; ; ; ; ; ; ;	+ (135 Xe)	+	: ; ; (135 Xe) ;		
 	2 2-a	+		+	+	+ (137 Cs, 140 Cs, 140 Ba)		+ (110 m Ag, 134 Cs, 136 Cs)
•	2'-b 2-c	- - - +	- : : + :		+	; ; ; ; ;		

Setting up accuracy requirements for these nuclear data is rather a matter of opinion subject to discussion, some arguments being :

- 1. large amounts of fuel failure make equipment maintenance extremely difficult and expensive if the plant is not designed with normal crud build-ups and fuel failures and the resulting high background radiation levels anticipated.
- 2. covering a factor of 2 incertitude by additional shielding is not expensive when designing the plant unless weight limitations as in large handling casks.
- 3. uncertainties in the mechanisms and parameters of fission product release and transport are always sufficient to-day to make predictions of coolant and reactor component contamination in error by a factor of 2.
- 4. people who are just trying to understand mechanisms and to measure parameters of fission product release and transport need small errors coming from the nuclear data side.

R.H. Flowers proposal is that we should be satisfied with fuel inventories calculated to within a factor of 2 leading to the following typical requirements :

- a) the cumulative yield to within a factor 1.5
- b) the half-lives of the nuclide and its significant precursors to within ± 10 %
- c) the neutron absorption cross-sections of the nuclide and its significant precursors with an accuracy which allows the term $(\lambda + \sigma \Phi)$ to be known within ± 10 % in total fluxes of 10^{14} and 5.10^{15} n.cm⁻².s⁻¹ for thermal and fast systems respectively.

Specifying that the errors quoted above are <u>two standard deviations</u> leads to yield requirements to 20 % standard deviation and other terms to 5 % standard deviation.

In addition to these accuracy requirements for fuel inventory calculations, γ -ray spectra (energy, abundance) must be accurate enough to permit a positive recognition of a given isotope in the presence of roughly equal amounts of the other contaminants or to enable gamma transport and dose rate calculations to within a factor 1.5 behind shields that attenuate the total dose by factors up to 10^3 .

TABLE 111-a

SHORT-LIVED FISSION PRODUCT DATA

			Cum	ulative Yield	(8)			Energy Emissic	n (MeV)
1502096	Harl-Lube ::	235 U _{th}	235 U 6	238 U 6	239 Pu _{th}	239 Pu	233 U _{th} :	е е	٢
85 m Kr	: . 4.43 <u>+</u> 0.06h	1.33 (8) ^{a/:}	1.43 (64)	0.81 (64)	0.60 (4)	0.64 (64)	2.21 (8)	0.29	0.15
87 Kr	:76.4 m	2.37 (16)	2.56 (64)	: 1.42 (64) :	0.95 (64)	1.11 (64)	3.09 (64);	1.35	0.74
88 Kr	2.8 ± 0.02 h	3.64 (32)	3.48 (64)	1.68 (32)	1.34 (64)	1.37 (64)	5.49 (64);	0.33	2.00
89 Kr		4.64 (8)	4.57 (64)	3.01 (64)	1.44 (8)	1.65 (64)	5.06 (8)	1.36	2.19
131 I	8.06 <u>+</u> 0.01 d	2.77 (4)	3.21 (4)	3.66 (8)	3.89 (2)	4.20 (4)	3.38 (4) :	0.18	0.37
133 I	:21.0 h	6.76 (4)	6.60 (32)	; 6.47 (32) ;	6.83 (4)	6.82 (32)	5.95 (4)	0.40	0.61
133 Xe	: 5.27 <u>+</u> 0.003d	6.77 (2)	6.60 (32)	. 6.47 (8)	6.84 (2)	6.82 (32)	5.96 (4)	0.10	0.08
[135 I]	: 6.70 h	6.39 (4)	6.30 (64)	5.67 (32)	6.04 (4)	6.29 (64)	4.75 (4) :	,	1
135mXe	: 15.8 m	1.05 (16)	1.01 (64)	0.85 (32)	1.06 (16)	1.90 (32)	0.95 (32);	•	0.53
135 Xe		6.72 (4)	6.45 (64)	5.75 (16)	7.22 (2)	7.45 (64)	6.04 (4) :	0.30	0.26
137 Xe	. 3.8 ш	5.94 (4)	6.14 (64)	: 5.95 (64)	6.05 (8)	5.78 (32)	5.90 (8)	1.72	0.14
						••	••	••	

a/ figures into brackets are relative mean standard deviation in % as quoted in NEDO 12154

T	11-94 414-	Branching			Cumula	tive Vield	(\$)		Energy E	nission (MeV)
14070pe	nach-eihe	ratio(4)	^{235U} th	235 U	238 U 6	239 Pu _{th}	239 Pu	233 U _{th}	β	Ŷ
85 ['] Kr	10.59 <u>+</u> 0.25 y		0.29 (1)	0.32 (4)	0.17 (32)	0.14 (2)	0.14 (4)	0.49 (2)	0.22	0.002
[90 Kr]	32.3 <u>+</u> 0.09 в		4.83 (4)	4.56 (64)	3.22 (64)	1.80 (64)	1.76 (64)	4.23 (8)	-	-
90 Sr	28.5 ± 0.8 y		5.94 (2)	5.24 (64)	3.28 (8)	2.12 (2)	2.09 (2)	6.48 (2)	0.17	0
[90 Y]		100							0.92	0.004
95 Zr	65.2 <u>+</u> 0.3 d		6.50 (2)	6.27 (4)	5.58 (4)	4.89 (2)	4.59 (4)	6.19 (2)	0.11	0,73
[95 ND]		100							0.05	0.77
103 Ru	39.6 + 0.2 d		3.11 (4)	3.22 (4)	6.40 (4)	6.97 (4)	6.53 (4)	1.80 (4)	0.06	0.49
[103 mRh]	-	99							0	0.04
106 Ru	369. + 3 d		0.39 (2)	0.97(16)	2.83 (8)	4,25 (4)	4.52 (16)	0.26 (2)	0.01	0
[106 Rh]		100							1.42	0.20
[109 Ag]	stable		0.027(16)	0.12(16)	0.27 (8)	1.38 (8)	1.62 (16)	0.039(16)	-	-
110m Ag	255. ± 6 d								0.07	2.67
[110 Ag]		13				į			1.18	0.05
125 Sb	2.73 + 0.11y		0.025 (4)	0.072(16)	0.11(16)	0.11(16)	0.19 (16)	0.11(16)	0.08	0.45
125 mTe]	-	21							0	0.14
129mTe	33.3 ± 0.1 d		0.10 (4)	0.50(16)	0.22(16)	0.28(64)	0.33(32)	0.29(64)	0.21	0.10
129 Tej		64			 				0.52	0.07
[133 Cs]	stable		6.77 (1)	6.60 (4)	6.47 (4)	6.84 (1)	6.82 (2)	5.96 (1)	-	-
134 Cs	2.09 ± 0.1 y								0.16	1.58
[135 CB]	3.10 ⁶ y		6.73 (4)	6.45 (4)	6.67 (8)	7.22 (2)	7.45 (2)	6.10 (4)	-	
136 Cs	13.65 ± 0.28 d		0.0068 (4)	0.0052(64)	0.011 (16)	0.10 (8)	0.15 (8)	0.082 (4)	0.10	1.86
[137 Xe]	3.8 m		5.94 (4)	6.14 (64)	5.95 (64)	6.05 (8)	5.78(32)	5.90 (8)	-	-
137 Cs	30.1 ± 0.5 y	100	6.23 (1)	6.19 (2)	5.95 (4)	6.53 (1)	6.62 (2)	6.56 (1)	0.17	0.
[137 m Ba]		93.5							0.	0.66
[140 Xe]	13.6 ± 0.1 s		3.83 (8)	3.16 (64)	5.59 (64)	1.73 (4)	1.65 (64	1.35 (8)	-	-
[140 Cs]		100							-	-
140 Ba	12.83 ± 0.04 d	100	6.30 (2)	5.90 (4)	5.95 (4)	5.49 (2)	5.14 (4)	6.13 (4)	0.29	0.15
[140 La]		100							0.50	2.23

TABLE 111-6 LONG-LIVED FISSION PRODUCT DATA

TABLE IV

.

NEUTRON CAPTURE CROSS SECTION FOR DIFFERENT REACTOR SPECTRA

:	CROSS SECTION ^{a/} (barn)									
: 1SOTOPE : :	$PWR = 2.5 \ 10^{13} = n.cm^{-2}.s^{-1}$		HTG Ø _{th} = 3 n	R .1 10 ¹³ .cm ⁻² .s ⁻¹	: LMFBR : $p_t = 4.3 \ 10^{15}$: $n.cm^{-2}.s^{-1}$:					
: : 135 Xe	4.66 1	0 ⁶ c/	2.10 1	0 ⁶ c/	: : : 0.					
: 109 Ag	670	c/	395	₫/	: : 0.747 c/					
: 110 m Ag ^{b/}	82	d/	: 82	d/	: 0.					
: : 133 Cs	194	c/	120	c/	: 0.51 c/ :					
: 134 Cs	140	d/	140 d/ :		: 0.					
: 135 Cs	29	d/	21	d/	: unknown					
:										
: a) for PWR and HTGR	a) for PWR and HTGR capture cross sections are given relative to the flux									
at 2200 m/s; thi	at 2200 m/s ; this flux is defined as yielding the actual absorption									
: Mate on 255 0 whe	rate on 235 U when using $\sigma_a = 680$ b									
b) the capture brand spectrum independ	b) the capture branching ratio between 110 m Ag and 109 Ag has been assumed spectrum independent and equal to 3.8 % [3]									
c) from [14]by avera	aging on the m	reactor	spectrum							
d) from [15] by usin	ng r = 0.3 in	PWR, r	= 0.2 in 1	HTGR						

Energy resolution of the kev order and intensities to within \pm 10 % for the few most intense γ -rays is sufficient. Gamma transport calculations need less accurate energy resolution but average group spectra to within \pm 20 % for group intensities ; typical group structures being as follows :

Δ	Е	=	100 KeV	between	0.1	and	0.5	MeV
Δ	Е	Ξ	250 KeV	between	0.5	and	1.5	MeV
Δ	Ε	Ξ	500 KeV	for E>	1.5	MeV		

In order to illustrate the incertitudes which occur in fuel inventory calculations, we have reported below three examples of such calculations corresponding to three typical power reactor conditions : PWR, HTGR, LMFBR.

Table III shows the nuclear data used : - cumulative yields come from Rider and Meek [13]

- the other data are taken from our library [3]

Neutron cross sections reported in Table IV were derived either by averaging point cross sections from Benzi [14] on reactor spectra or from Walker [15] thermal and resonance integral data ; in that case epithermal over thermal flux ratios were set equal to 0.3 for PWR and 0.2 for HTGR.

Capture branching ratios were taken from [15]. Although we have checked that for thermal reactors spectrum averaged cross sections from Benzi were consistent within 20 % with cross sections derived from Walker, we believe that the accuracy of the effective capture cross section is not better than \pm 30 % in thermal reactor. In fast reactor we have assumed an error of a factor 2.

The activity calculations have been made with the following assumptions concerning the irradiation conditions :

1. PWR : - irradiation time : 900 days
 - power distribution as function of irradiation time as follows (%) :

:	Period (day)	235 U	: 238 U :	: : 239 Pu :
:	0-300	76.5	: 6.5	: 17
:	0-600	65	: 7	: 28
:	0-900	55.5	: 7.5 :	: 37 :
:	900	30	: : 9 :	: : 61 :

2. HTGR : - irradiation time : 4 years - power distribution : 235 U 60 % 233 U 40 %
3. LMBFR :- irradiation time : 2 years - power distribution : 235 U 2.4 % 238 U 11.8 % 239 **Pu** 85.8 %

Results from calculations are given in Table V.

TABLE V-a ACTIVITY OF SHORT-LIVED FISSION PRODUCTS FOR DIFFERENT REACTOR TYPES

(Curie per Kilowatt)

ISOTOPE	85 m Kr	87 Kr	88 Kr	89 Kr	131 I	133 I	133 Xe	135m Xe	135 Xe	137 Xe
Reactor _{PWR} a/	6.8	13.	18	22	30	58	58	6.4	9.1	52
htgr	14.1	28	39	45	27	56	56	5.9	13.9	52
LMFBR	5.4	9.6	12.5	15	34	57	57	6.5	61	52
										_

a/ the activities quoted correspond to the composition of the fuel after 900 days irradiation.

TABLE V-b

ACTIVITY OF LONG LIVED FISSION PRODUCTS FOR DIFFERENT REACTOR TYPES IN

FUNCTION OF IRRADIATION TIME

(Curie per Kilowatt)

137 Cs 140 Ba	1.0 51	2.0 50	3.0 49	1.26 55	2.50 -	3.70 -	- 6.4	 0.63 47	1.25 -	1.86 -	
136 Cs	1.2	2.5	3°9	1.4	2.6	3.8	5.0	 0.9 ^{a/}	•	 I	
134 Cs	0.86	о • е	5.7	0.92	3.1	5.8	8.7	0.16	0.59	1.2	1
129 m Te	2.2	2.6	2.8	2.6	ł	1	1	2.5	1	1	
125 Sb	0.08	0.17	0.26	0.12	0.21	0.29	0.34	0.15	0.28	0.39	
110 mAg	0.010	0.043	0.088	0.001	0.003	0.005	0.007	0.005	0.015	0.030	
106 Ru	4.7	10.6	16	1.5	2.2	2.6	2.7	10	T7	22	
103 Ru	37	trti	48	23	1	I	I	 54	56)	
95 Zr	50	6 †	47	 54	56	I	ł	 35	0 11	+1 +	
4S 06	0.86	1.5	2.1	1.3	2.5	3.8	5.0	0.23	0.47	0.69	
85 Kr	0.12	0.21	0.29	0.21	0.40	0.59	0.76	 0.04	0.08	0.12	
ISOTOPE	30 0 d	PWR 600 d	900 q	1 Y	2 y	HTGK ³ y	4 V	0.5 y	1 y	LMFBR 1.5 y	

a/ direct contribution from fission only

*

The results in table V illustrate the sensitivity of activity calculations to different fissile species and different neutron spectra as present in different reactor types and in variable proportions as in PWR cores.

Some crude assumptions have been made in case of fast reactor fission yields :

- fissions in 241 Pu have been regarded as 239 Pu fissions
- the average of thermal fission and "fast" fission yields has been taken as "fast reactor" fission yield.

These assumptions plus the uncertainties of the basic nuclear data result in errors in the calculations that have been evaluated for both reactor types in the case of the most irradiated fuel.

The following statistical rules have been used :

- error on cumulative yields ;

wherever cumulative yields have been calculated by an expression of the form :

$$\bar{y} = \sum_{i} \alpha_{i} y_{i}$$

to account either for neutron energy or for repartition of fissions between different fissile species, the fractional standard deviation has

been estimated by :

$$\frac{\Delta \bar{y}}{\bar{y}} = \frac{\left[\sum_{i} \alpha_{i}^{2} y_{i}^{2} \left(\frac{\Delta y_{i}}{y_{i}}\right)^{2}\right]^{1/2}}{\sum_{i} \alpha_{i} y_{i}}$$

- overall error :

activity being calculated under the general form :

 $A = F (x_1, x_2, \dots, x_n)$

where the x_i 's are various nuclear data, the fractional standard deviation has been derived from the formulae :

$$\frac{\Delta A}{A} = \frac{\left[\sum_{i} \left(\frac{\partial F}{\partial x_{i}}\right)^{2} x_{i}^{2} \left(\frac{\Delta x_{i}}{x_{i}}\right)^{2}\right]^{1/2}}{F(x_{i})}$$

In case of fast reactor other incertitude factors may have been added :

a/ when significant difference was encountered between thermal (y_{th}) and "fast" (y_f) yields, the error was multiplied by :

$$\frac{\max (y_{th}, y_f)}{0.5 (y_{th} + y_f)}$$

b/ when contributions from 241 Pu fissions could modify the average yield value (on the basis of differences observed between thermal 239 Pu and thermal 241 Pu fission yields) another factor was added representing the effect of 20 % fission in 241 Pu.

Results from this analysis are presented in Table VI.

From Table VI we can draw the following conclusions, bearing in mind that all accuracies stated are one standard deviation :

- When known and if reliable the errors on half-lives have practically no effect; errors on half-lives should be obtained for 87 Kr, 89 Kr, 133 I, 135 m Xe, 137 Xe
- Error on short-lived isotope activity is due to cumulative yield error except for 135 Xe where it arises from capture cross section uncertainty; however these errors are acceptable.
- 3. The accuracy of long-lived isotope activity when directly produced by fission and not influenced by neutron capture is determined by yield error ; this error is in general lower than ± 10 % except for 125 Sb in fast reactors where both energy at which fission occurs and 241 Pu fissions introduce uncertainties and 129 mTe in all reactor types due to large errors on cumulative yields from 239 Pu and 233 U. Important gaseous precursors 90 Kr, 137 Xe, 140 Xe have accurate half-lives but not enough accurate yields for 239 Pu and in general for fast fission.
- 4. The error on 110 m Ag, 134 Cs and 136 Cs is in most cases governed by the incertitude on the neutron capture cross section of 109 Ag, 133 Cs and 135 Cs respectively ; the capture cross section of 135 Cs for fast reactor spectra is unknown. They are little influenced by errors in their own neutron absorption cross section because the term $\sigma \phi$ is not dominant in ($\sigma \phi + \lambda$). For fast reactors the 109 Ag yield is very much influenced by 241 Pu fissions ; in addition, the effect of neutron energy on the capture branching ratio is not known.

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TABLE VI

ERRORS ON ACTIVITY CALCULATIONS (%) (one standard deviation)

	:	: : P(WR	HTC	GR	: LMFBR		
Isotope	$\frac{\Delta \lambda}{\lambda}$ (%)	: : <u> </u>	total error	<u>∆y</u> y	total error	<u>∆y</u> y	total error	
85m Kr :	1	: 6	: 6	6	6	: 28	34	
87 Kr	U U	: : 33	: : 33	31	31	37	43	
88 Kr	<u>لا</u> ا	: : 31	31	36	36	37	42	
89 Kr	U	: : ⁹	: : 9	6	6	: 28	35 35	
131 I	< 1	2	: 2	3	3	2	5	
133 I	U	: 4 : 4	: 4	3	3	: 7	8	
133 Xe	< 1	: 4 :	: 4	: 3	3	. 7	8	
135mXe	U	: 11	: 11	16	16	20	20	
135 Xe	८ 1	2	: 30	3	30	29	29	
137 Xe	U	8	: 8	: 4	4	16	17	
85 Kr	2.3	: : : 1	: : 3	1	З	: : 12 : ¹²	: 12	
90 Sr	2.8	2	: 4	: 2	4	: . 7	: 8	
95 Zr	<1	2	: 2	: 2	2	: : 3	: 4 :	
103 Ru	<1	: 3	: 3	: 3	3	: : 3	6	
106 Ru	<1	: 3	: 3	2	2	: 8 : ⁸	12	
110mAg	2.3	(109) 8	: : 20	(109)11	20	(109)50	> 100	
125 Sb	4	11	: 12	: 12	13	50	50	
129mTe	<٦	50	: 50	: : 42	42	37	37	
134 Cs	4.8	(133) 1	: 26	(133) 1	25	(133) 2	100	
136 Cs	2	(135) 4	: 30	: (135) 3	30	(135) 3	> 100	
137 Cs	1.7	: 1	: 2	: 1	2	: : 2	3	
140 Ba	< 1	: 2 :	: 2 : 2	: 3	З	: 8 : 8	8	

It should be desirable to limit the errors for these three important fission products to ± 40 %.

5. Although no mention has been made of this, the eventual effect of other neutron reactions such as (n, p) and (n, 2n) reactions should be investigated.

Tritium production by ternary fission

This question has been left apart because tritium contamination although important in effluent release monitoring, has not been yet clearly identified as an important risk for plant personal health. However we will try to briefly situate the contribution of ternary fissions compared to other tritium sources in various power reactors.

As far as we know there has been much more tritium yield measurements for the thermal fission of 235 U [16-21] than for the other fission types [20, 21].

From the published 235 U ternary fission tritium yields considered equal weight we derived an average value of $(0.10 \pm 0.02) 10^{-3}$; the relative standard deviation for one measurement was found to be \pm 50 % which was arbitrarly used to characterize the incertitude of isolated yield measurement on other fissile species. Table VII shows the tritium yield values that we have used for estimating tritium generation in various reactors.

TABLE VII

TRITIUM TERNARY FISSION YIELDS

: : :	Fissile Isotope	:	Neutron Energy	: Tritium : yield :	Référence
:	235 U	:	thermal	$(0.10 \pm 0.02)10^{-3}$: (16) to : : (21):
:	238 U	:	2.5 MeV	. 0.14.10 ⁻³	: (20)
•	239 Pu	:	thermal	• 0.23.10 ⁻³	: (20)
:	239 Pu	:	1 MeV	. 0.25.10 ⁻³	(20)
:	241 Pu	:	thermal	0.26.10 ⁻⁴	: (20)
:	233 U	:	thermal	: 0.11.10 ⁻³	: (20)

In a PWR core at equilibrium with fission distribution 235 U (66 %) 238 U (7 %), 239 Pu (27 %) one obtains an effective tritium yield of $(0.14 + 0.04) \cdot 10^{-3}$.

The year tritium production by ternary fission in a 1200 MWe PWR would then be 19000 + 5000 Ci.

Assuming a 2 % release rate from the fuel to the coolant (1 % through zircalloy cladding plus 1 % from failed fuel) makes the fission contribution ($^{4}400$ Ci) still dominant compared to other tritium sources ($^{3}350$ Ci) from boron and lithium of the water.

However the error quoted above seems to be acceptable for tritium contamination evaluations inside the reactor building.

In a HTGR with fission distribution 235 U (60 %), 233 U (40 %), the effective tritium yield would be $(0.10 \pm 0.03) 10^{-3}$; however it is expected that ternary fission tritium production will not be the major source of contamination for the coolant, given low design fuel failure rates.

In a LMFBR on the contrary, ternary fission is the most important tritium source because of tritium diffusion through stainless steel cladding. From values in Table VII, effective yield should amount to about 0.2 10⁻³.

Although the actual uncertainty of this figure is not known and should be checked it is anticipated that the accuracy requirements will be more severe for environmental problems than for contamination problems for which an incertitude factor of 1.5 might be acceptable.

3 - FUEL FAILURE DETECTION

There are in fact two problems under this item :

- 1. the question of fuel failure detection which raises differently from a reactor type to another depending on safety arguments
- 2. the question of failed fuel location more related to contamination and environmental aspects.

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In gas-cooled reactors and LMFBR there must be a thorough surveillance of both bulk coolant activity and individual channels whereas in light water reactors and HTGR only the mean coolant activity is monitored.

The principle of burst can detection in gas-cooled reactors as R.H. Flowers says, is the measurement of krypton and xenon fission products released into the coolant gas, emphasis being placed upon isotopes of halflife smaller than the primary circuit circulation time in order to optimize channel discrimination. The gamma spectra must be known in sufficient detail to allow positive recognition of Kr and Xe isotopes of half-life greater than one second ; the cumulative fission yields of the gases should be known to within a factor 1.5, the half-lives to within ± 20 %.

The measurement of bulk coolant activity is used as an early warning of can failure. For this purpose, a long lived gas such as 133 Xe or 135 Xe or alternatively a group of neutron-emitting fission products is generally used. The delayed neutron precursors have the merit of giving good sensitivity in locations of high background γ -radiation ; an accuracy of \pm 20 % in fission yield of the gases and in group yields of delayed neutrons is quite sufficient for the purpose.

In a sodium-cooled reactor direct \checkmark -counting of the coolant is an insensitive method of detecting failed cans because \checkmark -emission from 24 Na is a very intense background ; however the possibility appears to be a counting of Kr and Xe stripped from the sodium immediatly after it leaves the channel ; the demands for nuclear data are rather similar to the gas cooled situation with the difference that individual and cumulative yields from typical mixtures of 235 U, 238 U, 239 Pu, 240 Pu, 241 Pu, 242 Pu and for typical fast reactor spectra should be known to within \pm 20 %. The isotopes of interest are 88 Kr, 89 Kr, 90 Kr, 91 Kr, 138 Xe, 139 Xe, 140 Xe, 141 Xe ; their half-life should be known with an error leading to less than \pm 20 % incertitude after up to 1 minute decay time. This arises from the need to know the threshold for detection for the safety argument.

Another possibility is a delayed neutron count on sodium from individual channels : K.W. Brindley points out that the delayed neutron detection system is establishing itself as an important safety channel for protection against sub-assembly faults at powers that are potentially hazardous. Transit times as low as 3 secondes are being considered, so delayed neutron precursors of half-life greater than 1 second, as those reported in Table VIII, are important for fast reactor fuel failure detection.

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TABLE VIII

: : : Isotope :	: : : Half-life :	: Neutron : : émission : : probability : : P _n (%) :
85 As	2.03 s	23 ± 3
87 Br	55.7 s	2.62 ± 0.05
88 Br	15.9 s	4.7 <u>+</u> 0.4
89 Br	4.55 s	8.8 <u>+</u> 0.9
90 Br	1.63 s	13 <u>+</u> 3.5
93 Rb	5.89 s	1.50 <u>+</u> 0.14
: 94 Rb	2.67 s	: 10.3 ± 1.3 :
137 I	24.4 s	5.1 <u>+</u> 1.0 :
138 I	6.8 s	1.9 ± 0.5
•	<u>.</u>	<u>.</u>

DELAYED NEUTRON PRECURSORS

All neutron yield data come from [23] except 85 As [22]. Although these neutron yields appear as rather accurate the fission yields of the isotopes are much uncertain according to [13] and it is anticipated that their half-life too has not the required accuracy.

The justification for asking data on individual neutron emitters rather than delayed neutron groups where more precise measurements and universal six periods sets seem to exist [24],could be a discrimination process during transport from fuel to counter between different precursor species. As far as we know, a detailed description of delayed neutron spectra is not necessary when counting is based on neutron slowing down followed by thermal flux measurement.

A new method of failed fuel location in course of implementation on fast reactors in based on tagging fuel elements by a unique isotopic mixture of noble gases [34] like 78 Kr, 80 Kr, 82 Kr or 126 Xe, 128 Xe, 129 Xe.

If a failure occurs, the analysis of the escaped gas tag identifies the ruptured element ; the method of analysis is in fact more complex because of the event of simultaneous failure ; in addition the effect of the flux on all gas ratios must be evaluated. Capture neutron cross sections for these isotopes are therefore required, especially for fast reactor spectra.

In HTGR, as pointed out by H.J. de Nordwall and J. Brisbois there is no necessity for a rapid fuel failure detection because the risk of fast and extended fuel melting is very unprobable. Detection of failed fuel requires only a relative measurement of coolant activity, preferably that associated with a single rare gas. For location of failed fuel, the proposed methods depend on recognition of a short-lived rare gas or delayed neutron emitters by an array of gas samplers. A short-lived activity is necessary to avoid confusion being caused by recirculation.

To design such a system, H.J. de Nordwall says, one must be confident that suitable isotopes will be released from the core and that changes in concentration can be recognized against the appropriate background Since the measurement is relative, precise decay schemes are unnecessary and half-lives within 20 % are probably adequate. The principal uncertainty is the efficiency with which the signal-generating nuclides can be attenuated by the fuel sleeves since this could determine sensitivity.

In light water reactors there is a tendancy towards real-time isotopic assay providing quantitative identification of isotopes important for both operational and effluent monitoring requirements, as those mentioned in part 2 of this paper. This would make use of on-line gamma-ray spectrometers with automated computer analysis and should require decay information for each isotope including gamma-ray energies to within 1 KeV, abundancies to within 10 % and half-lives to within 5 %. However, the use of radionuclide standards such as 137 Cs or 85 Kr could solve at the same time the problem of the detailed knowledge of the isotope gamma-ray spectrum and the question of detector calibration.

Failed fuel identification in light water reactors is presently done during refuelling outage by submitting each fuel element to the so-called "sipping" test which consists in measuring iodine isotopes released by failed rods into stagnant water. From experience and statistical evaluations people know the signal-over-background ratio to which one or more failed rods correspond: the need for nuclear data is then limited to those permitting to recognize the isotopes in question against the background.

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In conclusion, the requirements for fission product nuclear data for failed fuel detection and location could be summarized as follows :

- 20 % accuracy on half-lives and cumulative yields from typical fuel compositions and for relevant neutron spectra for noble gases : 85 m Kr, 87 Kr, 88 Kr, 89 Kr, 90 Kr, 91 Kr, 133 Xe, 135 m Xe, 135 Xe, 138 Xe, 139 Xe, 140 Xe, 141 Xe.
- 20 % accuracy on periods and yields of delayed neutron groups or preferably of individual delayed neutron precursors of half-life greater than one second from typical fuel compositions.
- 3. Capture cross section within 10 % for 135 Xe and stable noble gases that are used in gas tagging.
- 4. 5 % accuracy on half-life, 10 % on gamma-ray yields, gamma-ray energy resolution of the KeV order for intermediate and long-lived fission products important for contamination inside and outside the plant (see chapter 2).

4 - FISSION PRODUCT HEATING AFTER SHUTDOWN

We will confine ourselves to cooling times between 0 and about 10^7 seconds corresponding to the period of time between reactor shutdown and fuel transport towards reprocessing plants, the questions of spent fuel transportation itself, chemical processing and waste management being treated in other papers.

The knowledge of fission product energy release rate may be important in :

- reactor safety
- decay heat removal loops dimensioning
- fuel handling
- fuel storage

The relative importance and the accuracy requirements for the above topics are extremely reactor dependent.

In light water reactors, the assumption that a break occurs in the

primary coolant system piping would result in the rapid expulsion of a large fraction of the primary coolant into the reactor containment. Although the fission chain is stopped by loss of moderation, the fuel still generates about 7 % of the heat generated during power operation. The ability of the Emergency Core Cooling System to remove this heat in order to prevent clad melt and to garantee the core amenability to further cooling is evidently dependent upon the knowledge of fission product heating within the first few minutes after shutdown. H.J.de Nordwall points out that several BWR plants in the United States have recently been "derated" as a result of a new recognition that densification of UO2 fuel, early in fuel life, can affect the results of the Loss-of-Coolant Accident Analysis, causing somewhat higher peak cladding temperatures to be calculated. It would appear that some, if not all, of these deratings would have been unecessary if the allowance for uncertainty in fission product after-heat (an allowance which is included in the licensing criteria) were appreciably less than the + 20 % presently postulated in the criteria. For example, a 35 % increase in after-heat function would result in a BWR [25] in an increase of 192°_{r} in the peak fuel cladding temperature reached about 130 seconds after the onset of the accident. While relief might be found by reduction in uncertainties at several points in the analysis, it is undoubtedly true that an appreciable reduction in uncertainty in the afterheat function (e.g. to + 5 % or less) would contribute significantly to the avoidance of costly restrictions on power plant operations.

When analyzing accuracy requirements on fission product afterheat one must bear in mind the other sources of heat released after shutdown ; for a light water reactor this can be reduced to :

- residual delayed neutron fissions which depend on the magnitude of the negative reactivity at schutdown
- 239 U and 239 Np decay energy which depend on the fuel enrichment and neutron spectrum

Table IX illustrates the fractional contributions of the different heating sources in a three core region PWR at end of cycle with 4 % negative reactivity at shutdown [26].

From Table IX one can observe that the residual fission term is dominant within the first 10 seconds and it should be desirable to know, from reactor kinetics and delayed neutron group errors of paper 13, with which

TABLE IX

:	Time after shutdown (s)	: : :	Residual fissions	: : 2 :	39 U - 239 Np	•	Fission products
:	0.1	:	60	:	2	:	38
:	1	:	60	:	2	:	38
:	10	• :	49	•	3	:	48
:	100	:	0	:	10	:	90
:	1000	:	0	:	12	:	88

CONTRIBUTIONS TO AFTER HEAT FUNCTION IN A PWR (%)

accuracy this term can now be calculated for typical distributions of fissions as quoted in Table X.

The accuracy of the 239 U - 239 Np term is probably better than \pm 10 % and this has no effect on the total heating error. If we would assume that no errors come from the other terms, a \pm 10% accuracy on the total heating for example could be obtained with errors on the fission product heating term of \pm 25 % below 1 second, \pm 20 % at 10 seconds and \pm 10% at 100 seconds cooling time onwards.

TABLE X

DISTRIBUTION OF FISSIONS IN A PWR FUEL IN FUNCTION OF BURN-UP (%)

: : Bu : (Mu :	vin-up Id/T)	: : : :	235 U	:	238 U	:	239 Pu
: 1	1 000	:	63	: :	7	:	30
: 2	2 000	:	44	:	8	::	48
: 3	3 000	:	30	: : :	9	:	61

From table X it is apparent that both 235 U and 239 Pu afterheat functions must be known with the same accuracy while 238 U afterheat could be three times less accurate.

The theoretical approach to this problam consists in calculating the detailed inventory of fission product activities and then of individual energy releases starting from the largest number of fission product nuclear data. Even if we confine ourselves to the case of long irradiation times, which is the case of interest, where short-lived fissions products contribute less than for example in the case of a burst, present calculations underestimate the energy release because they dont take into account the energy from fission products with unknown nuclear data ; those with half-lives of the order of one second and less.

For example, Table XI shows a comparison between some recent calculations from Tasaka et al [28] and ourselves and the evaluation of gamma plus beta fission product energy release during reactor operation from James [30] for 235 U and 239 Pu thermal fission.

In the case of 235 U, calculations from England-Shure [33] and results of an ANS-5 Subcommittee work [31] based on a previous evaluation of Shure [32] have also been reported.

From this comparison it appears that calculations underestimate the energy release at zero cooling time by quantities that amount 4 to 24 % for 235 U and 9 to 34 % in the case of 239 Pu. If these figures could be confirmed, they almost should be considered as acceptable, as pointed out above.

The ten years old evaluation from Shure [32], based on old experimental measurements in that cooling time range, seems to ideally fill the gap between 0 and 1 second cooling time whereas it agrees with both evaluations to within 10 % for times ranging from 1 to 1000 seconds. The Shure's evaluation is given, for times below 1000 seconds with an incertitude of + 20 %, - 40 % which might seem pessimistic with regard to consistency of table XI-a results, but probably still reflects the wide dispersion of experimental results presently available. However, Tasaka and Sasamoto mention that their calculations of gamma energy release are consistent to within \pm 10 % with measurements between 10^{-2} and 10^3 seconds.

We therefore need bechmark afterheat experiments for 235 U and 239 Pu samples submitted to constant thermal fission rates during various times $(10, 10^2, 10^3, 10^4, 10^5 \text{ seconds})$ and considering cooling times from 0.1 to 10^5

TABLE XI-a

ENERGY RELEASED **BY 23**5 U THERMAL FISSION PRODUCTS AT SHORT TIMES AFTER SHUTDOWN (MeV/fission)

: Cool	: (ng : (s) :	England Shure	d : :	Devillers	: : Tasaka : Sasamoto	: Shure	: James
; ; 0	:		:		:	:	: :14.2 <u>+</u> 1.4
: 0.:	:		:	<u>,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,</u>	: : 12.2	: : 13.9	:
: : 1	:		:	11.5	: 11.3	. 11.8	•
: 10	:	9.3	:	9.2	: : 9.1	: : 9.9	:
: 100	0 :	6.6	:	6.1	: 6.3	: 6.7	:
: 100	00 :	3.9	:	3.6	: 3.7 :	: 3.5 :	•

(irradiation time 1 year)

TABLE XI-6

ENERGY RELEASED BY 239 Pu THERMAL FISSION PRODUCTS AT SHORT TIMES AFTER SHUTDOWN (MeV/fission) {irradiation time = 1 year}

:	Cooling time (s)	::	Devillers	:	Tasaka	James
:	0	:		::		: :12.5 <u>+</u> 1.6 :
:	0.1	:		:	9.8	:
: :	1	:	9.6	:	9.3	:
: :	10	::	8.1	:	8.0	: :
; ;	100	::	5.5	:	5.8	: :
:	1000	:	3.2	:	3.4	: :

seconds. The case of 241 Pu thermal fission should also be considered in the event of plutonium recycling. On the other hand we need evaluated data files including errors on individual data. Comparison of sensitivity calculations to experimental results would permit first to check the errors quoted on

data and secondly to determine whether or not it should be necessary to take into account new fission products or alternatively to derive heating functions, corresponding to lumped short-lived fission products, directly from measurements.

In our opinion, theoretical evaluations of fission product energy release have much progressed in the last few years thanks to new evaluations, compilations and measurements on fission yields, half-lives and Q values which have permit to increase the number of fission products that we can introduce into the calculations from about 300 when we were limited to half-lives greater than one minute to about 600 now, including half-lives of 1 second or less. We feel that given the large number of isotopes which contribute significantly to the heating function at short cooling times (roughly 20 chains including 100 fission products), a high accuracy on individual isotope energy release is unnecessary ; a factor of 2 would probably be sufficient to reach an accuracy of \pm 10 % on the total heating.

One of the most important field in our opinion where it could be paying to reduce the uncertainties is the fractional cumulative yields from 235 U, 239 Pu, 241 Pu thermal fission, for fission products of half-life lower than one hour and yield values above 1 %.

Concerning the half-lives it could perhaps be sufficient for the moment to evaluate their uncertainties.

As long as the knowledge of the energy distribution between beta and gamma radiations of different energies is not concerned in the design of ECCS, we need each isotope decay scheme just enough detailed as to permit a calculation to within 30 % of the mean neutrino energy E_v it emits per desintegration, unless nuclear models reveal to meet this requirement.

The effective energy release per isotope decay is then calculated by $E = Q - E_v$, Q being the decay energy which should be known to within ± 10 %.

The other factor which might influence the heating function is the effect of the neutron flux ; this effect has been checked using the methods and data described in [1] [3] and the fission product neutron capture cross sections from Walker [15] and wherever possible spectrum averaged cross sections derived from Benzi [14].

Table XII illustrates the magnitude of the effect in function of cooling time for 235 U and 239 Pu irradiated 900 days.

TABLE XII

EFFECT OF NEUTRON CAPTURE ON 235U AND 239 PU THERMAL FISSION PRODUCT ENERGY RELEASE

(irradiation time = 900 days ; thermal flux = $2.5 \ 10^{13} \ n.cm^{-2}.s^{-1}$

: : : Cooling : time (s) :	Fraction of	235 U { operating ver	: Deviation:	2 Fraction o p	: : :Deviation:	
	Without capture	With capture		Without capture	: With : capture :	
: : 10 ⁰	5.97 ⁻²	6.00 ⁻²	: : : : : : <1 :	4.86 ⁻²	: : 4.92 ⁻²	: 1
: 10 ¹	4.77 ⁻²	4.80 ⁻²	: <1 :	4.08 ⁻²	: : 4.14 ⁻²	: 1
: 10 ²	3.16 ⁻²	3.17 ⁻²	: <1 :	2.80 ⁻²	: : 2.82 ⁻²	: 1
: 10 ³	1.86 ⁻²	1.87 ⁻²	: <1 :	1.65 ⁻²	1.67^{-2}	: 1 :
: 10 ⁴	9.17 ⁻³	9.27 ⁻³	: 1 :	8.01 ⁻³	: 8.19 ⁻³	: 2
: 10 ⁵	4.30 ⁻³	4.48 ⁻³	: 4 :	4.00 ⁻³	: 4.35 ⁻³	: 6
: 10 ⁶	2.18 ⁻³	2.31 ⁻⁹	: 7 :	2.01 ⁻³	: 2.18 ⁻³	: 9
: 10 ⁷	6.36 ⁻⁴	6.94 ⁻⁴	: 9 : : 9 :	6.57-4	: 7.17 ⁻⁴ :	: 9 :

The energy released from fission has been assumed 30 192.9 MeV for 235 U and 198.5 MeV for 239 Pu.

The effect of neutron captures appears to be negligible within the first hours after shutdown, reaches 5 % after one day cooling time, when fuel unloading takes place, and 10 % after three months cooling time, at the end of the fuel storage period. These figures are upper bounds of the effect because we have considered the most irradiated fuel. On the other hand, the effect of the flux in that range of cooling times is only due to 134 Cs generation through capture. The error on that term due to an assumed 30 % error on the capture cross section may be evaluated to 26 % (from results of chapter 2), which introduces at 10^7 seconds cooling time an uncertainty of 2 % on the total heating of 235 U fission products (this error could reach 5 % after 1 year cooling time). For 239 Pu, the situation is very similar. For cooling times between 10^2 and 10^7 seconds, we do think that sensitivity calculations starting from errors (to be evaluated) on individual data would be able to provide a good garantee on theoretical calculations ; as paper 15 will show, the confusing situation of experimental integral data does not give any mean for deriving error factors to be applied to the calculations. Special mention must be made however of calorimetric measurements by Lott et al [35] which have had the merit of providing the total energy release from 235 U thermal fission products. These measurements seems to indicate that calculations are accurate to within 10 % for cooling times between 100 and 10^7 seconds. Such experiments should be repeated in other laboratories and extended to the 239 Pu and 241 Pu cases.

In the case of gas cooled reactors as K.W. Brindley points out, thermal reactor specialists **accomodate** to uncertainties in decay heat such as ± 20 %. In the loss of coolant accident, the sensible heat in the fuel, which amounts to 11 full power seconds in AGR's, dominates the safety **i**ssue so only a low accuracy on decay heat is required.

In HTGR, Gulf General Atomic Company has investigated the sensitivity of their core-cooling analysis to uncertainties in decay heat [36]. They find that HTGR cores, because of their large heat capacity, are relatively insensitive to uncertainties in the afterheat function. They compared calculated peak temperatures versus delay time before startup of the Core Auxiliary Cooling System (CACS) following a loss-of-Main-Loop-Coolant Accident, for their nominal after-heat function and for an alternate function that was 20 % higher than nominal for t $\leq 10^3$ sec. and 10 % higher than nominal for t $> 10^3$ sec. The time within which the CACS must be started up in order that no critical safety temperature limits should be exceeded was reduced from about 40 minutes to about 26 minutes. This is probably not a really significant difference, since there is no question that the CACS must function sooner of later. Nonetheless, H.J. de Nordwall feels that it would probably be helpful to HTGR designers if the uncertainties were reduced to ± 5 %. The opinion of J. Brisbois is that an accuracy of ± 10 % at cooling times from 10 minutes to a few days could be sufficient for HTGR designing.

Remembering that in HTGR cores 60 % of fissions are produced in 235 U, and the remainder in 233 U makes at cooling times of 100 seconds onwards at least 60 % of the afterheat accurate t_0 within \pm 10 % as observed previously. Concerning the after-heat resulting from 233 U thermal fissions, different theoretical evaluations agree to within \pm 10 % between 10² and 10⁷ seconds as paper 15 will show. However no integral measurement exists that can warrant this

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TABLE XIII

CONTRIBUTION OF DOMINANT 233 U FISSION PRODUCT TO AFTER- HEAT (%)

Irradiation time:	4 years
Operating power :	1 watt
Cooling time :	10 ³ seconds

1

ISOTOPE	BETA HEATING 0.5475 10 ¹¹ MeV/s	GAMMA HEATING 0.6554 10 ¹¹ MeV/s
87 Kr	2.9	-
88 RD	8.2	2.4
88 Kr		5.2
89 Sr	2.4	
91 Sr	2.4	2.1
91 Y	2.3	
92 Sr	-	3.8
92 Y	5.6	-
93 Y	4.8	-
94 Y	4.4	
95 ¥	2.0	
95 Zr	2	2.3
95 Nb/	-	2.4
97 Zr	2.3	-
132 I	-	5.2
134 I	2.2	6.6
135 I	_	3.9
138 Xe	-	2.4
138 Cs	3.6	5.9
139 Ba	3.1	-
140 La	-	7.6
142 La	3.2	7.1
143 La	2.1	-
144 Pr	3.3	_
Rest	39	39

accuracy ; here again calorimetric measurements should be profitable. Turning to the sensitivity aspect, let us attempt to derive an estimation of the error of the after-heat calculation for 233 U thermal fission, as an example of what could be done if we had error files available. Let us consider for example the fission product heating after a 4 years irradiation of 233 U and a decay time of 10³ seconds corresponding roughly to the first cooling time of interest for HTGR. The total heating function is the sum of individual heating **from** about 50 isotopes ; Table XIII shows the contribution of the most important of them (contributions greater than 2 % of either beta or gamma heating have been reported).

The following simplified assumptions have been made for estimating the error :

- errors of half-lives are negligible against the others
- errors of cumulative yields are those quoted by Meek and Rider [13]
- errors of the energy emitted by each isotope is assumed constant and equal to \pm 20 %, resulting for example from \pm 10 % on the desintegration energy Q and \pm 30 % on the mean neutrino energy E_V.
- the total error is calculated as

with

$$\frac{sP}{P} = \frac{\left(\sum_{i} s^{2}_{Pi}\right)^{1/2}}{P}$$
$$\frac{s_{Pi}}{Pi} = \left[\left(\frac{s_{Yi}}{Yi}\right)^{2} + \left(\frac{s_{Ei}}{Ei}\right)^{2}\right]^{1/2}$$

Pi being the heating due to isotope i, Yi its cumulative yield and Ei the energy emitted per decay of isotope i (correlations between isotopes belonging to the same chain have been accounted for)

The error calculated as indicated above is \pm 11 %; this figure must of course be considered as illustrative because of the crude assumption we have made but it gives an idea of the order of magnitude the uncertainties in data should reach to meet the accuracy requirements. Based on more reliable errors such calculation would provide good estimates of calculational errors.

The effects of neutron captures on the after heat functions have been checked as shown in Table XIV, by the same method as used in the case of PWR.

TABLE XIV

EFFECT OF NEUTRON CAPTURE ON 235 U AND 233 U THERMAL FISSION PRODUCT ENERGY RELEASE

Irradiation time : 4 years Thermal flux : $3.1 \ 10^{13} \ n.cm^{-2}.s^{-1}$

		235 U		233 U				
: Cooling : time : (sec)	Fraction (poi	of operating ver	: :Deviation: :	Fraction o p	: :Deviation :(%)			
: : :	Without capture	With capture	: {\$} : : : :	Without capture	: With : capture :	: (6) : : : :		
10 ⁰	5.98 ⁻²	6.02 ⁻²	1	5.00 ⁻²	5.14 ⁻²	ζ1		
101	4.78 ⁻²	4.82 ⁻²	<1	4.40 ⁻²	4.43 ⁻²	Հ۱		
: 10 ²	3.17 ⁻²	3.18 ⁻²	<u>الا</u> م	-2 3.14	3.15 ⁻²	<u>۲</u>		
10 ³	1.87 ⁻²	1.88 ⁻²	<u>کا</u>	1.92 ⁻²	1.94 ⁻²	1		
104	9.25 ⁻³	9.40 ⁻³	2	9.68-3	9.84 ⁻³	2		
: 10 ⁵	4.37 ⁻³	4.54 ⁻³	: 4 : 4	4.45 ⁻³	4.65 ⁻³	: 4		
: 10 ⁶	2.25 ⁻³	2.41 ⁻³	: 7	2.27 ⁻³	2.40 ⁻³	6		
: 10 ⁷	7.01-4	7.74-4	10	6.56-4	. 7.31 ⁻⁴	. 11		

The energy released from fission were 192.9 MeV for 235 U and 190 MeV for 233 U.

Again the effect is due entirely to 134 Cs with similar conclusions as in the case of PWR concerning its influence on the error.

In fast breeder reactors the accuracy requirements, as expressed by K.W. Brindley are the following :

- propagation of melting in sub-assemblies requires an accuracy of ± 20 % in decay heat for decay times of 1 minute onwards.
- failure of all forced circulation requires data to calculate the start-up of natural convection to a lower accuracy (± 30 %) for 1 minute onwards because of uncertainties in the hydraulics.

- decay heat removal loops are sized on the decay heat arising several hours after shutdown, after allowing the sodium temperature to rise. The integrated decay heat is therefore important and it is required to an accuracy of ± 15 % over 0 to 24 hours for pool reactors
- reactor fuel handling requires an accuracy of ± 10 % for transfer of fuel from the core to a store outside of the reactor from 8 hours onwards. Penalties for having to extend this because of errors in decay heat arise because of the cost of replacement electricity and also the extra fuel inventory. In the long term it might be possible to justify ± 5 % accuracy.

Here we are faced with a new problem which is the influence of neutron energy on fission yields from 239 Pu and 241 Pu, the latter contributing up to 20 % of the fissions in future stations.

First of all we have examined this influence by comparing after heat from 239 Pu thermal fission to after heat from 239 Pu "fast" fission using the relevant yields from Meek and Rider.

This influence has been found to be 1 % or less in all cases; similar results have been obtained by K. Tasaka et al [28]. The effect of neutron energy can therefore be considered as smaller than the calculational errors in the case of 239 Pu. For 235 U it would be higher ; for 241 Pu it cannot be evaluated.

As paper 15 will show, recent calculations of 239 Pu afterheat are consistent to within 10 % between 10^2 and 10^7 seconds cooling time, for long irradiation time. Integral experiments are scarce; some confirm beta heating calculations at short times to within 10 % and more often 5 % like measurements of Mac Nair [37] whereas others show desagreements as large as factor of 2 in the gamma heating [38]. The present lack of measurements and the dispersion of the available ones does not permit to warrant any accuracy although we feel that we are not far from \pm 10 % for the decay times considered.

The effect of neutron capture has been evaluated as shown in table XV by only accounting for the isotopes considered in the cross section evaluation of Benzi [14].

As in thermal reactors the effect is due to 134 Cs ; an incertitude of a factor 2 in the capture cross section of 133 Cs would for example introduce an error of 4 % in the afterheat at 10⁷ seconds.

TABLE XV

EFFECT OF NEUTRON CAPTURE ON 239 Pu FAST FISSION PRODUCT ENERGY RELEASE

Cooling time (sec)	23 Fraction of Po Without capture	239 Pu Fraction of operating <u>Power</u> Without : With capture : capture				
: : : 10 ⁰	: : : : : : : : : : : : : : : : : : :	4.83 ⁻²	< 1			
: : 10 ¹	: 4.05 ⁻² :	4.07 ⁻²	: <1 :			
: 10 ²	: 2.79 ⁻² :	2.80 ⁻²	: <1 :			
: 10 ³	: 1.64 ⁻² :	1.65 ⁻²	<1 <1			
104	7.90 ⁻³	8.00 ⁻³	: 1 :			
10 ⁵	4.03 ⁻³	4.11 ⁻³	2			
: 10 ⁶	1.95 ⁻³	2.00 ⁻³	: 3 :			
: 10 ⁷	6.17 ⁻⁴	6.40 ⁻⁴	: 4 :			

Irradiation time : 2 years Total flux :4.310¹⁵n.cm⁻².s⁻¹

In conclusion, the accuracy requirements related to fission product after-heat, as expressed by designers, can be synthetized as follows :

- only long irradiation times are of interest : this makes the knowledge of very short lived fission products not so stringent (irradiation times of 10⁵ seconds or such are infinite for them)
- 2. fissile species of primary interest are :

233 U thermal fission235 U thermal fission239 Pu thermal fission

3. of secondary interest are :

238 U fast fission 241 Pu thermal fission 239 Pu and 241 Pu fast fission in the event of non negligible effect of neutron energy contrary to that has been observed for 239 Pu

- 4. the decay times of interest start from 0 for 235 U and 239 Pu, from 10^3 seconds for 233 U
- 5. an accuracy of + 10 % should be reached at each decay time of interest with + 5 % as a long term aim.
- error files are required for cumulative yields, half-lives, branching ratios, desintegration energies, neutrino energies, capture cross section of 133 Cs in order to evaluate calculational errors
- 7.to check the calculational errors, benchmark experiments (calorimetric measurements if possible) are requested especially for decay times below 10⁵ seconds.

Acknowledgments

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Appendix:

LISTS OF FISSION PRODUCTS IMPORTANT FOR AFTERHEAT

case	table	fissile	neutron	MeV per	irradiation
	no.	isotope	spectrum	fission	conditions
A1	A-I	235 _U	thermal	192.9	900 days, thermal flux:
A2	A-II	239 _{Pu}	thermal	198.5	2.5 x 10^{13} n.cm ⁻² .s ⁻²
A3	A-III	233 _U	thermal	190.0	4 years, thermal flux:
A4	A-IV	235 _U	thermal	192.9	3.1 x 10^{13} n.cm ⁻² .s ⁻²
A5	A-V	239 _{Pu}	fast	198.5	2 years, fast flux: 4.3 x 10 ¹⁵ n.cm ⁻² .s

The contributions of important fission products to the total afterheat have been calculated for the following 5 typical cases:

The total power was 1 watt in all cases. In the following tables fission products that contribute more than 1% to total afterheat are listed in descending order of importance for cooling times of 1,10,10², ..., 10⁹ seconds. Since the headings of the computer printouts are in French, their English translation is given below:

temps de refroidissement - cooling time puissance - power contributions a la puissance residuelle totale -- contribution to total residual power

4

------TEMPS DE REFROIDISSEMENT= 0.1000E 01S T Ŧ I Ĩ PUISSANCE TOTALE= 0.3752E 12MEV/S I T PUISSANCE BETA = 0.1946E 12MEV/S I PUISSANCE GAMMA = 0.1806E 12MEV/S T Ĩ --- 1 7 t I CONTRIBUTIONS A LA PUISSANCE RESIDUELLE TOTALE (%) I 1 T 2.1 ----*
 VUCLIDE
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 P.BETA
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 P.GAMMA
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 I
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 94
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 2.11
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 1.09
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 I
 CS140
 I
 2.07
 I
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 I
 CS140
 I
 2.00
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 1.444
 I
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 I
 T134F
 I
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 I
 CS138F
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 I
 0.99
 I
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 I
 CS138F
 I
 1.87
 I
 0.64
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 PA 90F
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 LA142
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 T NUCLIDE I P.TOTALE I P.BETA I P.GAMMA I TRESTE(%) I 53.31 I 29.44 I 23.87 I

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Ţ	NUCLIDE	I	Ρ.	TOTA	LE	I	P.86	TA	I	P.GAMMA]
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Ţ	Y 96	Ţ		2.5	4	I	1.	30	I	1.24]
I	CS140	Ţ		2.4	6	I	1.	29	Ţ	1.17	1
I	T134F	Ţ		2.3	9	I	C.	52	I	1.86]
Ţ	CS138F	I		2,3	7	Ĩ	C.	80	I	1.56]
I	RB 91	I		2.3	5	I	1.	69	1	0.66]
Ţ	RB 90F	I		2.3	3	I	1.	05	I	1.28	J
Ī	I136F	I		2.2	8	1	1.	01	J	1.27	1
Ţ	LA142	I		2.0	6	I	C.	56	Ī	1.50]
I	LA140	Ţ		1.9	5	I	0.	34	I	1.61	J
Ţ	RB 89	I		1.7	5	I	C.	57	Ŧ	1.17	1
Ţ	XF138	J.		1.7	lą.	I	C.	36	I	1.38	1
I	SR 92	Ĩ		1.7	2	T	٢.	78	I	0.94	1
I	KR 89	I		1.7	2	I	€.	66	I	1.06	1
I	Y 94	Ĩ		1.7	1	I	1.	31	Ι	0.40	1
Ţ	C\$139	I		1.5	9	I	1.	12	I	0.47	1
Ţ	LA144	I		1.5	6	I	1.	02	I	0.54]
I	Y 95	I		1.4	7	Ĩ	l.	14	Ŧ	0.32	1
Ţ	RB 88	Ţ		1.3	7	I	1.	01	I	0.36]
I	I137	I		1.3	7	here	0.	57	Ţ	0.80	1
I	1135	I		1.2	8	I	C.	27	I	1.01	1
Ţ	KR 90	Ţ		1.2	7	I	C.	51	T	0.76	J
I	SB132F	I		1.2	2	I	С.	55	I	0.66	1
Ţ	XF137	I		1.2	Ú.	I	1.	11	I	0.09]
I	1132	I		1.1	8	I	C.	23	I	0.95]
I	PP 88	I		1.1	8	I	C.	61	I	0.57	1
I	SR 95	I		1.1	8	I	<u>.</u> ,	43	I	0.75	1
I	TE133F	I		1.1	7	Ţ	С .	45	I	0.72	1
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LA140	1		2.0	96		I		Ç	• 1	52		Ĩ	2.44
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Y 94	I		2.	52		I		1	•	34		I	0.59
Y 96	I		2.4	45		I		1	2	26		I	1.19
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KR 89	I		10	38		ĩ		C	•	72		I	1.16
1132	t		1.	79		I		C	• 3	35		I	1.44
1136F	I		1.	78		¥		C	•	79		I	0.99
TE133F	I		1,	71		I		C		56		I	1.05
Y 92	Y		1.	59		1]	• 4	45		I	0.25
MC101	Ţ		1.	54		¥.		C	•	32		I	1.32
CS140	I		1.	54		I		Ç		31		I	0.73
TC172F	I		1.	48		I		C	•	73		I	0.55
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Ţ	TEM	25	DE REFI	ROIDI	SSE	ENT=	0.10	OCE O	55	
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٤	140	I	10.	11	I	1.	77	I	8.34	
1	132	I	6.	05	ľ	1.	17	I	4.88	
۲	92	I	5.	02	Ŧ	4.	29	T	0.73	
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١	1 93	I	3.	86	I	3.	57	I	0.29	
NE	97F	I	3.	59	ľ	1.	46	I	2.13	
1	[133	I	3.	56	I	la	42	I	2.14	
S F	2 91	I	3.	46	I	1.	69	I	1.77	
1	[134F	ĩ	3.	41	I	C.	75	I	2.66	
P	2144F	I	3.	37	I	3.	29	I	0.08	
ι <i>ι</i>	142	I	3.	09	I	C.	.84	I	2.24	
25	95	I	3.	18	I	C.	40	I	2.68	
NE	95F	I	2.	91	I	Ç.	17	I	2.74	
ZF	2 97	T	2.	81	I	2.	.09	I	0.72	
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)	91F	I	1.	97	I	1.	96	I	0.01	
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I	LA140	I	23.	53	Į		3.	60	I	16.93]
I	I 132	I	10.	13	I		1.	96	I	8.17	1
I	PR144F	I	6.	94	I		6.	78	I	0.17	1
I	ZR 95	I	6.	29	I		C.,	81	I	5.48	1
Ţ	NB 95F	Ţ	6.	02	I		C .	35	I	5.67	, 1
I	Y 91F	I	4.	34	I		4.	02	I	0.02	I
I	I 133	I	3.	23	I		1.	29	I	1.94	1
I	PA140	Ι	3.	16	I		1.	98	I	1.18	1
I	MO 99	Ĩ	3.	35	I		2.	02	I	1.02	1
I	SP 85	I	3.	33	I		3.	03	I	0.0	Ĩ
Ĩ	CF143	I	2.	81	I		1.	49	I	1.32	1
Ţ	NB 97F	1	2.	69	I		1.	10	I	1.60]
Ţ	PR143	T	2.	14	I		2.	14	I	0.0	1
Ţ	ZR 97	τ	2.	7 ר	Ι		1.	54	I	0.53	1
I	RU1/13	Ţ	1.	91	I		с.	22	I	1.69	I
ĩ	FU156	I	1.	62	I		٥.	38	I	1.24	1
Ţ	1131	Ţ	1.	60	1		с.	53	I	1.07	1
Ţ	CE141	I	1.	51	1		1.	04	I	0.48	I
Ţ	Y 93	I	1.	44	I		1.	33	I	9+11	1
I	<u>NB 97M</u>	Ţ	1.	40	I		с.	0	I	1.40	1
I	XF133F	I	1.	36	I		Ċ,	75	I	0.61	1
I	CS134F	Y	1.	33	I		С.	12	I	1.21	J
Ţ	TE132	I	1.	26	I		e.	23	I	1.03	1
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	10L		• ۲ مد هده د			- L.					τ¤ ••••••	-	۰ مــــــــــــــــــــــــــــــــــــ	r •0^	•
LA14	0	I		24	• 57	2	I		4	4.	30		I	20.	22
PR14	4F	I		13	.1	5	ſ		12	2	84		I	0.	32
NB 9	5F	I		11	. 5!	5	1		0	2	66		I	10.	88
ZR 9	5	1		10	.94	4	I		1	L	41		T	9.	53
Y 9	1 F	I		6	.9	5	ľ			5.	91		I	0.	04
SP 8	9	I		5	.13	3	I		5	5.	13		I	0.	0
EA14	ņ	I		3	.5)	I		1	2.	19		I	1.	30
RU10	3	Ĩ		3	.10)	I		().	36		I	2.	74
PR 14	3	I		2	. 60)	I		Ż	2	60		I	0.	0
CS13-	4F	I		2	• 51	7	I		(3.1	24		I	2.	33
CE14	1	I		2	• 3	7	I		1	L.	62		I	0.	75
113	2	I		2	.12	2	I		(2•4	41		I	1.	.71
EU 15	6	I		1		5	I		(].	46		I	1.	49
113	1	I		1	•2	7	I		().	42		I	0.	85
CF14	4	I		1	. 1 (5	I		(2.	86		I	0.	30
PH17	6F	I		1	•14	4	I]	1.	00		I	0.	14
ND14	7	I		1	•03	2	I		(]•	58		I	0.	,44
REST	 F(%)	 T			. 9/	 <	 T			,	 26		 T		1

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I	TEM	PS	DE	REFR	101	SSE	MENT	r =	C. :	1000	08	5	
Ţ													
Ţ	PUI	SSI	ANCE	TOT	ALE=	0.	4330	5E	10	MEV/S	5		
Ī	PUI	SS/	ANCE	BET	4 =	0.	2480	DE	10	MEV/S	5		
Ι	PUI	SS4	ANCE	GAMI	4A =	- Q.	1850	5E	10	MEV/S	j -		
:			-							~~~~			
1	CONTRIOU	T T T	-	4 1 4			Nor	05	C 1 1		с т.		/ w \
L T	CUMENTOU	111	3N2	ALA	FUI	334	INCE	R.C	210	DUELI		JIALC	141
1													
T	NUCLIDE	T	D.	τοτλι	F	т	D	85	TΛ			D . C A MA	ι 1 Λ
			ге 		 	*) ۲۰ سرمین میں میں ا		1 ~ .	1 ** ** ** ** **	. 1	• • • • • • • • • •	
Ţ	PR144F	T		33.9	3	ĩ	-	33.	11	T		0.81	
Ť	NP 95F	Ī		19.9	5	ī	-	1.	15	1		18.81	
Ŧ	78 94	Ī		11.9	8	Ť		1.	55	Ĩ		10.44	•
Ī	CS134F	Ŧ		7.8	Ś	Ī		ō.	72	I		7.08	3
Ţ	Y 91F	I		6.7	7	I		٤.	73	1		0.04	ł
Ţ	SR 89	Ţ		4.2	5	I		4.	25	1		0.0	
I	RH106F	I		3.1	1	I		2.	73	I		0.38	3
Ţ	CE144	I		2.9	9	I		2.	22	1	•	0.77	,
I	Y 90F	I		2.3	2	I		2.	31	3		0.01	
I	RU103	I		1.6	5	I		с.	19	I		1.47	•
Ţ	EA137M	I		1.5	6	I		C.	0	1	•	1.56	>
2-			• ~ ~ ~										
Ŧ	RESTE(%)	I		3.6	5	I		2.	23	I		1.43	5
: -				-									

****** TFMPS DE REFROIDISSEMENT= 0.1000E 095 Ŧ Ŧ Ţ T PUISSANCE TOTALE= 0.4914E 09MEV/S Ţ I PUISSANCE BETA = 0.2890E 09MEV/S I T PUISSANCE GAMMA = 0.2024E 09MEV/S Ŧ Ť - 2 T T I CONTRIBUTIONS A LA PUISSANCE RESIDUELLE TOTALE (%) I I Ŧ T NUCLIDE I P.TOTALE I P.BETA I P.GAMMA I

 I
 CS134F
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 27.92
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 I ______ - - -_____ I RESTE(3) I 0.87 I 0.66 I 0.21 I

I TEMPS DE REFROIDISSEMENT= 0.1000E 105 T I T PUISSANCE TOTALE= 0.9826E 08MEV/S I T PUISSANCE BETA = 0.6473E 08MEV/S I Ţ PUISSANCE GAMMA = 0.3352E 08MEV/S Τ T - : I T T CONTRIBUTIONS A LA PUISSANCE RESIDUELLE TOTALE (%) I Ŧ T T NUCLIDE I P.TOTALE I P.BETA I P.GAMMA I I Y 90F I 47.25 I 47.02 I 0.23 I I EA137M I 33.45 I 0.0 I 33.45 I I CS137 I 9.45 I 9.45 I 0.0 I I SR 90 I 8.86 I 8.86 I 0.0 I TRESTE(%) I 0.99 I C.55 I 0.44 I •

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Ţ	TEM	PS	Đ€	REFR	DIC	DISSE	MENT=	0.1	CODE C	15	I
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Ţ	PUI	55/	ANCE	BET	A	= 0.	1586E	121	EV/S		Ĩ
Ţ	PUT	SSI	NCE	GAM	MΔ	= 0.	1490F	121	EV/S		Ī
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T											7
Ť	CONTRIBU	T 1 1	2 M		DI	17554	NCE DE	STO		TOTALE 191	2 T
Ŷ	CONTRIDU	. 16	. 11.5	M 1. M	fι	ACCIL	NUC NC	210		ICIALE 161	a T
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ः ज	11245	1		202	ງ າ	1 *		20	1 *	Le (O	i *
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1	1514	1		201	0	I -	le	10	1	1.00	1
1	¥ 96	<u>}</u>		1.9	1	I	с.	98	I	0.93	ĺ
}	(S138F	1		1.8	8	I	C.	64	I	1.25	1
Į	M01/33	I		1.8	7	I	0.	49	I	1.37	I
T	LA140	I		1.6	3	Ţ	¢.	29	Ţ	1.34	Ĩ
Į	10142	I		1.6	3	1	C.	44	1	1.18	ĩ
Ţ	TC 104	1		1.6	1	I	C.	72	Ī	0.89	I
Ţ	NB100F	T		1.6	0	I	1.	19	I	0.41	I
T	1137	Ŧ		1.4	6	I	С.	61	I	0.86	Ī
Ţ	TC102F	I		1.4	4	I	C.	91	ĩ	0.53	ĩ
I	F132	Ţ		1.4	0	I	C.	27	ſ	1.13	I
Ŧ	SP 95	I		1.3	6	Ţ	C.	50	1	0.86	I
Ţ	CS139	Ī		1.3	5	I	C.	95	I	0.40	1
ĩ	XE138	T		1.3	2	I	C.	28	I	1.04	ĩ
Ţ	M0101	I		1.3	0	ſ	Ċ.	26	I	1.04	I
T	1135	Ţ		1.2	9	T	Č.	27	Ţ	1.92	ĩ
Ī	TC106	Ţ		1.2	8	Ť	1.	09	Ţ	0.19	ĩ
Ţ	TC105	Ţ		1.2	7	Ť	ō	37	r	0.91	Ŧ
Ţ	TE135	Ĩ		1.2	6	Ĩ	6.	47	Ť	0.84	ĩ
T	\$8132F	Ĩ		1.2	5	Ť		57	Ī	0.68	Ť
Ţ	XE137	ĩ		1.2	ĩ	Ť	1.	11	ĩ	0.09	Î
Ŧ	RR 92	1		1.1	â	ĩ	с. С.	£7	1	0.57	7
ĩ	Y 95	Ť		1.0	ģ	ı T	с. С.	ρ <u>ς</u>	Ţ	0.24	7 7
Ŧ	¥ 94	Ť		1.0	â	Ť	n ve	83	f	0.25	л. Т
Ţ	NO135	T		1.0	e e	T T	• •	34	ı T	0.74	1 7
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r put	SSA	NCF	- B	ET.	Δ_	-	0.	12	34	E	1:	2 M F	V.	/S				
r PUI	SSA	NCE	ĒĞ	AM	MA	H	0.	12	901	Ē	1	2 M E	EV.	15				
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CONTRIBUT	r r c	INS	A	LA	ρ	UI	SSA	NCI	ΕI	RE	S	101	.EI		T	CTAL	.E	(8)
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NUCLIDE	I	Ρ.	. TO	TA	LE		I	(P., I	BE	T/	4		Ŧ		P.GA	MM	A
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CS140	I		2	• 3	3		Ι			1.	27	2		Ĭ		1.	11	
CS138F	I		2	• 2	4		Ĩ		(0.	70	5		I		1.	48	i
r y 96	I		2	, 1	9		I			1.	1	2		I		1.	07	,
E MOIDA	I		2	<u>。0</u>	2		Y		(۵.	5	3		Ĩ		1.	48	i
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E MO101	I		1	• 5	4		I		I	с.	3()		Ţ		1.	24	•
I T135	I		1	. 5	4		I		I	¢.	32	2		ĩ		1.	21	
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XE137	Ī		ĩ	.4	2		Ţ			1.	3	1		Ĩ		0.	11	
1 137	I		l	.3	7		I		(С.	5	7		I		0.	80	1
1 TC106	I		Į	• 3	4		Ĩ			1.	1	4		Ĩ		0.	20	ł
Y 95	I		1	.3	0		1			۱.	0	1		I		0.	29	ł
Y 94	Ŧ		1	• 2	9		I		4	C .	9	9		Ī		0.	30)
I SR 95	Ţ		1	•2	7		Ţ		1	0.	4	6		Ĩ		0.	81	
I SR 93	Ĩ		Ĩ	•]	8		I			C.	5	3		I		0.	65	i
I TF133F	I		I	•]	6		I		ŧ	0.	4	5		I		0.	72	
FB 91	I		1	•]	5		ĩ		i	Ċ.,	8	3		Ĩ		0.	32	
1 10144	I		1	.1	4		I		ł	с.,	7	4		I		9.	39)
PA141	I		1	.1	1		I		ł	r,	7	2		I		0.	. 39	1
1 10105	I		1	.1	0		I		1	c.	3	5		I		0.	.76)
I NR 98F	I		1	• 0	8		I			1.	0.	2		I		0.	.06	•
TF135	I		1	• ^	7		ĩ		ļ	Ċ.	3	6		I		0.	71	
I XE139	Ţ		Į	°J	2		I			0.	7	8		I		0.	24	ł
: I RESTE(%)	 F		49	.7					2	 7.	2	0		 I		22.	51	
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T PUT	SS/	ANCE	TOT	ALE	= 0.	17656	1	2MEV	115	
t PUI!	SS/	ANCE	BET	Α	= 0.	82526	1	1 MEI	115	
I PUI :	SSI	ANCE	GAM	MA	= 0.	94035	1	1 M E V	1/5	
*							and the second second			ويور حوية بريان مركة الرور ويور الأبر بران برياد بريد بالأبر الري
T										
I CONTRIBU	TI	ONS /	A LA	PU	HSSA	NCE R	ES	IDLE	ELLE	TOTALE (%)
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TNUCLIDE	I	P • 1	TOTA	LE	I	P.8	ET	A	I	P.GAMMA
*****		~								
I 1134F	I		3.9	1	I	C	a 8:	6	I	3.05
I CS138F	I		3.2	7	I	1	• 1	1	I	2.16
I LA140	Ι		2.8	4	I	Ç	• 5	0	I	2.34
T LA142	I		2.9	3	I	9	•7	7	I	2.06
I TC174	Ι		2.7	3	I	1	• 2.	2	I	1.50
I I132	Ţ		2.4	.4	I	C	• 4	7	I	1.97
1 TC192F	T		2.2	9	I	1	, 4	4	I	0.85
I I135	J		2.2	5	I	0	a 4	7	I	1.77
T CS139	I		2.1	6	Ţ	1	• 5	2	I	0.64
1 XE138	I		2.1	2	I	C	. 4	4	T	1.68
<u>I MO101</u>	Ī		2.1	1	I	C	. 4	2	I	1.70
I Y 96	I		2.0	4	I	1	• 0	5	I	1.00
1 10105	I		2.0	0	I	C	• 5	7	Ι	1.43
T Y 94	I		1.8	5	I	1	. 4	2	I	0.43
T T136F	I		1.8	2	I	C	• 8	1	I	1.01
J Y 95	I		1.7	'5	I	1	• 3	7	I	0.39
T XE137	I		1.6	6	I	1	. 5	4	I	0.13
I TE133F	I		1.6	3	Ĩ	C	• 6	3	I	1.00
T 80141	I		1.5	5	Ι	1	.0	0	I	0.55
* SR 93	I		1.5	50	I	C	. 6	8	¥	0.82
1.05140	I		1.3	7	I	C	.7	2	I	0.65
1 SP132F	Į		1.3	3	I	C	• 6	1	I	0.72
I \$8131	I		1.2	9	I	<u>c</u>	.3	8	I	0.91
J J133	I		1.2	3	I	C	. 4	9	ĩ	0.74
1 PR146	I		1.2	2	I	C	.4	0	Ι	0.82
T NB 97F	Ţ		1.1	4	I	C	. 4	6	I	0.67
T M0103	Ţ		1.0	8	I	C	.2	9	I	0.79
T PB 90F	Ī		1.0	6	Ĩ	Ċ	.4	8	Ī	0.58
I LA141	ĩ		1.0	5	Ī	1	.0	2	ī	0.03
1 80105	Ţ		1.1	4	Ī	Ċ	.3	9	Ī	0.65
I RHINGE	I		1.0	2	Ī	Č	. 9	0	I	0.13
T RESTE(%)	Ţ		42.4	3	I	22	.3	2	I	20.11

Ţ TEMPS DE REFROIDISSEMENT= 0.1000E 045 Ŧ T Ŧ PUISSANCE TOTALE= 0.1044E 12MEV/S 1 1 T PUISSANCE BETA = 0.4483E 11MEV/S I PUISSANCE GAMMA = 0.5958E LIMEV/S I I :--Ŧ T T CONTRIBUTIONS A LA PUISSANCE RESIDUELLE TOTALE (%) I Ĩ T • 2 ************
 I
 NUCLIDE
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 I NUCLIDE I P.TOTALE I P.BETA I P.GAMMA I I RESTE(%) I 25.15 I 11.80 I 13.35 I _____ ***************

TEMPS DE REFROIDISSEMENT= 0.1000E 055 Ĩ T I 1 PUISSANCE TOTALE= 0.5120E 11MEV/S I T PUISSANCE BETA = 0.2183E 11MEV/S T I PUISSANCE GAMMA = 0.2937E 11MEV/S Ĩ Ŧ 2 -Ŧ T I CONTRIBUTIONS A LA PUISSANCE RESIDUELLE TOTALE (%) I I ĩ _____ _____ 2-I NUCLIDE I P.TOTALE I P.BETA I P.GAMMA I

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 LA140
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 1.03 I
 0.23 I

 ¥ 92 0.39 Ţ I I I PR144F I I ZR 95 I I Y 93 I 0.06 I 2.22 I I Y 93 I I N8 95F I 0.19 Ĩ 2.27 I I 0.07 1 2.39 2.32 I 2.20 ./M I 1.94 CE143 I 1.88 I EU156 I 1.81 I RB 88 I 1.58 I SR 91 I 1.57 I BA140 I 1.56 I SR 92 I 1.34 I I131 I 1.7 I SB129 I I TE132 I LA141 1.49 I 2.05 I 0.74 I 1.94 I 0.88 I 1.38 I 0.41 1 0.80 Ĩ 0.58 I 1.14 Ŧ 0.87 1 0.85 Ĩ 0.85 I I RESTE(%) I 18.73 I 10.36 I 8.38 I

•

Ī	TEM	PS	DE F	REFRO	IDIS	SE	IENT	=	0.	1000E	065	: I
T T	PUT	ss/	NCE	TOTA	LE=	0.2	2717	E	11	MEV/S		I
Ī	PUT	ss.	NCE	BETA	=	0.1	1094	E	11	MEV/S		Ī
I	PUI	ss/	ANCE	GAMM	IA =	0.1	623	E	11	MEV/S		I
: •		~										******
I T T	CONTRIBU	T I (INS A	LA	PUIS	ISAN	CE	RE	SI	DUELLE	TOTALE	(%) I (%) I I
Ĩ.	NUCLIDE	I	P.1	INTOTAL	.E	I	Ρ.	BE	TA	I	P.GAMM.	A I
Ţ	LA140	I	1	18.nc)	I		3.	17	I	14.91	 I
I	1132	I	1	12,56)	I		2.	43	I	10.13	I
Ţ	RH106F	Ţ		6.51		I		5.	71	I	0.80	I
I	PR144F	I		4.82	2	ĩ		4.	70	Ţ.	0.12	I
Ţ	ZR 95	I		4.75	5	Ĭ		Ç,	61	1	4.14	I
Ţ	NB 95F	I		4.54	•	Ľ		٥.	26	I	4.28	I
T	RU103	t		4.30	}	I		C •	50	ri 1	3.80	I
I	1133	I		3.27	,	Ĩ		1.	30	I	1.97	I
Ţ	EU156	I		3.25	5	I		c.	77	ĩ	2.48	I
Į	NC 99	I		3.20)	I		2.	13	I	1.07	Ĩ
I	FA140	¥		2.79	7	I		1.	75	I	1.04	I
Ī	NB 97F	I		2.50)	Ĩ		1.	02	Ĩ	1.48	I
Ţ	1131	Ţ		2.25	i	1		C .	75	I	1.50	ľ
I	CF143	I		2.09	}	I		1.	11	I	0.98	I
Ţ	<u>7R 97</u>	I		1.92		I		lo	43	I	0.49	I
I	Y 91F	I		1.66	,	I		1.	65	I	0.01	I
1	PP143	1		1.59	,	I		le	59	ĺ	0.0	Ĩ
I	1-132	I		1.56	7	I		C.	28	I	1.28	I
1	CE141	Ĩ		1.54	} •	ľ.		1.	96	ĺ	0.49	I
1 *	XE133F	ľ		1.37	•	i T		Ge	16	Į,	0.61	I
1		1		1055	•	1 T		Ue n	12	1	1.20	l
I. T	SR 89	l		1.05	5	i T		90 10	0 05	I	1.30	I
: -]	RESTE(%)	 1	1	11.73	3	I		6.	<u>ç</u> 9	I	5.64	: I
¥ *								-				

÷ --TEMPS DE REFROIDISSEMENT= 0.1000E 07S I I I Ţ I PUISSANCE TOTALE= 0.1362E 11NEV/S - 1 1 PUISSANCE BETA = 0.5845E 10MEV/S I PUISSANCE GAMMA = 0.7778E 10MEV/S 1 I - : T T CONTRIBUTIONS A LA PUISSANCE RESIDUELLE TOTALE (%) I T T I ***** I NUCLIDE I P.TOTALE I P.BETA I P.GAMMA I

 I LA140
 I
 22.18
 I
 3.89
 I
 18.29
 I

 J RH106F
 I
 12.74
 I
 11.17
 I
 1.57
 I

 I PR144F
 I
 9.37
 I
 9.15
 I
 0.23
 I

 I N8 95F
 I
 8.94
 I
 C.51
 I
 8.43
 I

 I ZR 95
 I
 8.47
 I
 1.09
 I
 7.38
 I

 I RU103
 J
 7.15
 I
 0.83
 I
 6.31
 I

 I FU156
 I
 4.03
 I
 C.96
 I
 3.08
 I

 I FU156
 I
 4.03
 I
 C.96
 I
 3.08
 I

 I FU156
 I
 4.03
 I
 C.96
 I
 3.08
 I

 I FA140
 I
 3.16
 I
 1.98
 I
 1.18
 I

 I S134F
 I
 2.67
 I
 C.52
 I
 2.17
 I

 I C5134F
 I
 2.67
 I
 C.52
 I
 2.42
 I</ \mathbf{r} a set where the set of th I RESTE(%) I 7.53 I 3.52 I 4.01 I TEMPS DE REFROIDISSEMENT= 0.1000E C85 T E Ŧ 1 I PUISSANCE TOTALE= 0.448CE 1CMEV/S I PUISSANCE BETA = 0.2727E 10MEV/S PUISSANCE GAMMA = 0.1753E 10MEV/S T I Ŧ - 1 ---- 2 Ŧ T I CONTRIBUTIONS A LA PUISSANCE RESIDUELLE TOTALE (%) I Ŧ T 2. - 1 I NUCLIDE I P.TOTALE I P.BETA I P.GAMMA I

 I
 FH176F
 I
 31.82
 I
 27.91
 I
 3.91
 I

 I
 PR144F
 I
 22.11
 I
 21.57
 I
 0.53
 I

 I
 PR144F
 I
 22.11
 I
 21.57
 I
 0.53
 I

 I
 PR144F
 I
 22.11
 I
 21.57
 I
 0.53
 I

 I
 PR195
 I
 14.13
 I
 C.81
 I
 13.32
 I

 I
 ZR 95
 I
 8.49
 I
 1.09
 I
 7.39
 I

 I
 CS134F
 I
 7.42
 I
 0.68
 I
 6.73
 I

 I
 RU173
 I
 3.51
 I
 0.41
 I
 3.10
 I

 I
 Y
 91F
 I
 2.62
 I
 2.60
 I
 0.02
 I

 I
 CE144
 I
 1.94
 I
 1.455
 I
 0.50
 I

 I
 EA137M
 I
 1.54
 I
 C.0
 I
 1.54< 2. ****** T RESTE(%) I 5.04 I 2.95 I 2.09 I

----T. TEMPS DE REFROIDISSEMENT= 0.1COCE 095 Ŧ T T PUISSANCE TOTALE= 0.5639E 09MEV/S Ţ T PUISSANCE BETA = 0.3354E 09MEV/S ï I PUISSANCE GAMMA = 0.2285E 09MEV/S I T • Ŧ T I CONTRIBUTIONS A LA PUISSANCE RESIDUELLE TOTALE (%) I 1 Ι I NUCLIDE I P.TOTALE I P.BETA I P.GAMMA I

 I
 PH196F
 I
 35.35
 I
 31.00
 I
 4.35
 I

 I
 CS134F
 I
 23.89
 I
 2.20
 I
 21.69
 I

 I
 PR144F
 I
 13.84
 I
 13.51
 I
 0.33
 I

 I
 PR144F
 I
 13.84
 I
 13.51
 I
 0.33
 I

 I
 PR144F
 I
 11.46
 I
 0.33
 I
 I

 I
 PR137M
 I
 11.46
 I
 0.033
 I

 I
 PA137M
 I
 11.46
 I
 0.03
 I

 J
 Y
 90F
 I
 5.82
 I
 5.79
 I
 0.03
 I

 I
 CS137
 I
 3.24
 I
 3.24
 I
 0.00
 I

 I
 EU154
 I
 1.81
 I
 C.277
 I
 1.54
 I

 I
 CE144
 I
 1.22
 I
 0.91
 I
 0.01
 I

 J
 SR 90
 I *------------------------- 2 I RESTE(%) I 2.29 I 1.48 I 0.81 I ----******* _____

TEMPS DE REFROIDISSEMENT= 0.1000E 105 Ŧ I I ŧ. PUISSANCE TOTALE= 0.6323E 08MEV/S I T PUISSANCE BETA = 0.2900E 08MEV/SŦ T PUISSANCE GAMMA = 0.3423E 08MEV/S ĩ Ţ. $= = a_1 + a_2 + a_3 + a_4 +$ I T I CONTRIBUTIONS A LA PUISSANCE RESIDUELLE TOTALE (%) I Ŧ T I NUCLIDE I P.TOTALE I P.BETA I P.GAMMA I I EA137M I 52.88 I C.O I 52.88 I I Y 9CF I 25.68 I 25.56 I 0.12 I I CS137 I 14.94 I 14.94 I 0.0 I I SR 90 I 4.82 I 4.82 I 0.0 I I EU154 I 1.30 I 0.19 I 1.11 I _____ -----------. . . -----I RESTE(%) I 0.38 I 0.35 I 0.04 I ·
Table A-III: ²³³U thermal fission, case A3

ΞĘ TEMPS DE REFERDIDISSEMENT= 0.1000E 015 T Ŧ I PUISSANCE TOTALE= 0.3210F 12MEV/S r Ŧ PUTSSANCE RETA = 0.1636E 12MEV/S T Ŧ PUISSANCE GAMMA = 0.1574F 12MEV/SŤ T . T T I CONTRIBUTIONS A LA PUISSANCE RESIDUELLE TOTALE (%) I T T I NHELIDE I P.TOTALE I P.BETA I P.GAMMA ŧ

 1
 RR 01
 I
 P.1004(P)
 I
 P.8014
 I
 P.0484

 I
 RR 01
 I
 2.45
 I
 1.77
 I
 0.69

 I
 RR 91
 I
 2.44
 I
 0.80
 I
 1.64

 I
 KR 89
 I
 2.32
 I
 0.88
 I
 1.43

 I
 RB 90F
 I
 2.27
 I
 1.02
 I
 1.24

 I
 C\$140
 I
 2.17
 I
 1.14
 I
 1.03

 I
 1.412
 I
 2.17
 I
 1.14
 I
 1.03

 I
 1.412
 I
 2.17
 I
 1.14
 I
 1.03

 I
 1.412
 I
 2.17
 I
 1.14
 I
 1.39

 I
 1.345
 I
 2.11
 I
 0.71
 I
 1.39

 I
 1.36F
 I
 2.01
 I
 0.89
 I
 1.50

 I
 A140
 I
 1.92
 I
 0.34
 I
 1.59

 T T T Ţ Ţ Ţ Ŧ T Ŧ - Ŧ T T Ŧ T Ŧ Ŧ Ţ Ţ ۳ T I KR 9R I T I132 I Y 95 T I KP 90 I I XF137 I I LA144 I I I137 I I Y 92 I Ţ T T T Ŧ Ţ I Ţ T NRIOOF T Ţ I 1135 I T : -n and a star with the star wit I RESTE(*) I 50.19 I 27.17 I 23.01 I ******

:									-	:
t	ТЕМ	05	DF P	FERN	TDISSE	MENT=	0.	10005	025	Ţ
Ţ										1
Ţ	PHT	\$5	ANCE	TOTA	1 E= 0,	2770F	12	MEV/S		1
T	phi	55	ANCE	BETA	≂ ∩,	1381F	12	MEV/S		1
T	PUT	\$5	ANCE	CAMM.	$\Lambda = 0,$	1389F	12	MEV/S		Į
:				-						
Ţ										r
Ť	CONTRIBU	TT	ONS A	LA	PUTSS	NCE R	FSI	DUELLE	TOTALE (*) [
Ŧ	•			-						T
T	NUCL TOF	۲	P.T	OTAL	F T	P. 81	FTA	Ţ	P-GAMMA	Ţ
•		·			· · ·					
- T	DB 80	Ŧ		2.83	۲	0	. 03	Ţ	1.90	Ŧ
*	D 8 01	, Y		2 61	T	1	<u>ຊ</u> ຊ	T	0.73	*
T	VD 90	, T		7 61	י ד	1	00	T I	1 6 1	ŗ
1 7		1 T		2 60	ו ד	1	17	L T	1	1 T
ז ד	1 8127	I T		7 . 14	, T	1.	۰۱، ۵۵	l T	1 + 47	7
4 	1 HT 47	i T		7 11	ז ד	~ ~	117 07	r T	1471	1 7
I T	1 31 38F	1		2 - 44	T T		• · · · • • •	1	1+01	1 *
1	05140	ł		7.37	1	1.	1.1	1	1.10	,
1	кн жи	1		2.77	1	1.	.66	1	0.54	,
1	LAL40	1		2.23	1	0.	20	E -	1.84	1
1	1 (34F	1		2.20	1	о. •	48	1	1.72	1
Ĩ	1136F	1		2.19	Ţ	9.	. 07	1	1.77	Ĩ
1	Y 95	1		7.14	1	1.	-/14 	1	1.04	}
3	¥ 94	1		1.97	1 -	ι.	,47	1	0.45	
I	SR OX	1		1.72	1	<u> </u>	,78	Ţ	0.04	1
I	XF138	1		1.60	I	0.	33	J	1.27	1
Ŧ	CS139	Ŧ		1.59	3	1.	12	I	0.47	T
I	KB 88	T		1.53	Ŧ	<u> </u>	,??	Ţ	1.31	Ţ
I	1132	I].52	Ţ	Λ.	,29	T	1.23	T
Ţ	88 98	Ţ		1.51	Ţ	Α.	78	Ţ	0.73	Ŧ
Ŧ	Y 95	Ţ		1.49	Ţ	1.	16	Ţ	0.33	Ţ
I	RP 86	Ţ		1.48	Ţ	Λ.	74	T	0.75	Ť
Ŧ	RF 87	I		1.48	I	0.	<u>81</u>	1	0.66	Ţ
Ţ	XF137	I		1.36	Ť	1.	25	T	0.10	Ī
I	Y 92	I		1.31	Ţ	1.	12	Ţ	0.19	Ţ
T	1135	I		1.22	ĭ	ο.	26	I	0.96	Ţ
ĭ	L 1144	T		1.21	τ	0.	79	T	0.42	T
Ŧ	KB OU	T		1.18	Ĩ	0.	47	I	0.70	ĩ
T	SP 02	T		1.11	T	Λ.	16	T	0.95	Ŧ
T	RR 90M	T		1.09	T	٥.	37	I	0.72	Ŧ
I	SR132F	T		1.06	T	<u> </u>	48	T	0.58	Y
7	KR 87	T	-	1.05	I	n.	65	Ţ	0.40	T
Ţ	R4141	Ţ		1.04	Ţ	0.	67	T	0.37	T
I	1137	Ţ		1.04	Ţ	<u></u>	43	T	0.61	Ţ
T	Y 93	Ţ	•	1.03	Ţ	<u>^</u>	95	Ť	0.08	Ţ
: -								·		
Ī	PESTE(%)	Ţ	4	1.56	Ţ	22.	23	Ţ	19.33	Ţ
-	۰ ۲۰۰۰ - ۲۰ ۵۰ «۱۰۸۰ «۱۰ ۵۰ ۵۰ مور دو «۱۰ ۱۰ وور س								······································	

TEMPS DE REFRAIDISSEMENT= 0.1000E 035 T T T Ţ Ŧ PUISSANCE TOTALE= 0.1970E 12MEV/S T T PHISSANCE BETA = 0,933PF 11MEV/S Ŧ PUISSANCE GAMMA = 0.1037E 12MEV/S T T ٠ T I CONTRIBUTIONS & LA PUISSANCE RESIDUELLE TOTALE (9) T T T

 INICLIDE
 I
 P.TOTALF
 I
 P.RETA
 I
 P.GAMMA

 I
 RR R9
 I
 3.91
 I
 1.28
 I
 2.62

 I
 A142
 I
 3.48
 I
 0.95
 I
 2.53

 I
 CSI38F
 I
 3.41
 I
 1.15
 I
 2.62

 I
 R8
 R1
 3.17
 I
 2.34
 I
 0.83

 I
 IA14C
 I
 3.14
 I
 0.55
 I
 2.59

 I
 I134F
 I
 3.07
 I
 0.67
 I
 2.40

 R8
 90F
 I
 2.65
 I
 1.01
 I
 1.64

 I
 Y 04
 I
 2.62
 I
 2.01
 I
 0.51

 I
 KR 8R
 I
 2.14
 I
 0.41
 I
 1.72

 I
 KR 8R
 I
 2.09
 I
 0.44
 I
 1.66

 I
 Y 95
 I
 1.92
 I
 50
 I
 0.42

 I
 T NUCLIDE T P.TOTALE T P.BETA I P.GAMMA Ţ 1 T J T 2.59 Ţ T Ŧ T T Ţ T Ţ T Ŧ Ţ Ţ T T Ŧ T Ţ Ţ T Ĩ Ŧ Ţ T T Ţ T Ţ Ŧ T
 RA142
 I
 1.04
 I
 0.39
 I
 0.65
 I

 1133
 I
 1.91
 I
 0.40
 I
 0.60
 I
 Ţ T RA142 Ţ : -____ _ _ _ _ . ----- 2 T PESTE(9) I 32.15 I 16.62 I 15.53 I ******

TEMPS OF REFPOIDISSEMENT= 0.1000E 04S T Ŧ ۲ I PUISSANCE TOTALE= 0.1212E 12MEV/S T PUTSSANCE RETA = 0.5485E 11MEV/S 7 Ţ PUISSANCE GAMMA = 0.6640F 11MEV/S T T T I CONTRIBUTIONS A LA PUISSANCE RESIDUELLE TOTALE (") T T _____ 1 -T NHELTDE I P.TOTALE I P.BETA Y P.GAMMA I

 I
 NHCLIDE
 I
 P.TOTALE
 I
 P.BETA
 I
 P.GAMMA

 I
 A142
 I
 5.30
 I
 1.45
 I
 3.85

 I
 LA142
 I
 5.30
 I
 1.45
 I
 3.85

 I
 LA140
 I
 5.10
 I
 0.90
 I
 4.21

 I
 RR 8R
 I
 5.01
 I
 3.70
 I
 1.32

 I
 GS138F
 I
 4.85
 I
 1.64
 I
 3.21

 I
 I134F
 I
 4.50
 I
 1.64
 I
 3.21

 I
 I134F
 I
 4.50
 I
 1.64
 I
 3.21

 I
 I134F
 I
 4.50
 I
 1.64
 I
 3.21

 I
 I132
 I
 3.47
 I
 0.67
 I
 2.80

 I
 I132
 I
 3.47
 I
 0.67
 I
 2.80

 I
 I133
 I
 2.08
 I
 2.54
 I
 0.43

 و و به موجه بیدی میچه موجه موجه موجه به موجه به موجه به موجه موجه موجه موجه به موجه بود موجه بود موجه موجه بود و و T T Ţ 1 Ţ ۲. Ţ Ţ - 1 Ţ T T Ţ T T Ť Ţ Ţ T Ŧ Ţ Ţ Ţ 1.37 I 0.35 1.02 I ſ 1 78 07 I 1.35 1.27 Ť T NR OSF ĩ T 0.08 T 1.27 T 1.19 T 0.45 T 0.82 Ţ T 84141 T 0.92 0.26 T T Y 05 T 0.92 I 0.26 0.62 I 0.55 0.43 I 0.65 1.06 I 0.0 0.75 I 0.31 0.22 I 0.92 1.03 I 0.01 0.30 I 0.72 0.52 J 0.49 Ţ 1.17 1.08 1.06 I CF143 I I BR 84F I I SP 89 I Ţ Ţ T Ĩ Ŧ I 1.06 T 1.04 T Ţ 1 (5139 T 5 T TF133M T I YOIF I 1.04 T Ĭ 1.02 I 1.02 I 0.72 0.49 Ţ I SRI31 Ţ I TEISLE I F د * ها بین موجد این موجد بین موجد بین موجد موجد بین موجد بود بود بود بود بود بود موجد بود موجد بود بود بود بود بود * I RESTE(?) I 21.22 I 9.62 I 11.60 Ŧ

:							
Ţ	TEM	Þ۶	NE REFRAIDI	SSF	-MENT= 0.100	00E	055
T T	DUIT	cć	ANCE TOTALE-		6140E 11ME	115	
т Т	601 1011	33. CC	ANCT BETA -	- 0.	1014-0 11MC	110	
! T	DUT	יר כ כ כ		· · ·	3217E 11 MEN	110	
т +	-01	. د ق		· · · •			
ŗ							
Ţ	CONTRIBU	ΤTI		551	NICE RESTOU		TOTALE (7)
Ť	(., ,,		5	
:							
Ţ	NUCL TOF	T	P. TOTALE	T	P.BETA	I	P.GAMMA
:				•• •• •			······································
1	1 / 1 40	1	10.05	1	1.(5	ļ	9.79
1	1132	1	6.68	1	1.79	1	5.34
1	RR RR	1	5.54	1	4,08	1	1.45
1	Y Q2	1	5,09	Ĩ	4.35	1	0.74
I	1135	Ţ	4.11	I	0.87	ł	3.25
F	YQZ	I	3.87	T	3.58	T	0.29
T	SR 0]	Ţ	3.60	T	1.76	I	1.94
Ţ	KP RR	Ţ	3,47	Ţ	0.49	T -	2.48
Ŧ	1 1142	I	3.17	J	0,87	Ī	2,2()
T	NR 07F	I	3.12	I	1.27	I	1.85
1	PP]44F	Ĩ	5,02	٢	2.05	1	0.07
I	1133	1	2.99	Ţ	1.10	1	1.80
Ţ	78 05	l	2.82	ł	0.36	1	7.45
J	NR 95F	Ţ	2.66	Ĭ	0.15	Ţ	2.53
T	SR 92	Ţ	2.47	I	0.36	<u> </u>	2.11
I	ZR 97	T	2.44	Ţ	1.81	I	0.62
T	1134F	Ţ	2.32	Ŧ	0.51	I	1.81
Ţ	(F143	T	2.20	Ť	1.17	1	1.03
Ţ	SP 89	Ť	2.10	I	2.10	1	n.n
Ĩ	LA141	Т —	2.06	I	2.01	1	0.06
ľ	v ojf	I	2.05	ſ	2,04	T -	0.01
۲ -	NR 97M	Ţ	1.65	ł	0.0	I	1.65
1	HA]4()	Ţ	1,58	T T	n, oo	1	(1,59
ſ	MD DQ	1	J.49	ſ	0,99	<u>र</u>	0,50
Ţ	Y Q1M	Ţ	1.25	T	n_n	Ĩ	1.25
Į	KP 87	Ţ	1.04	Ţ	0.65	1	0.39
ĺ	1131	1	1.02	Ĩ	(1, 14	ļ	0.68
1	PR143	1	1.00	i	1.00	Ĭ	0.U
: · 1	RESTE(%)	I	15,13	Ī	7.13	ſ	8.01
						-	

Ţ	TFM	P۶	DE REEPOID	ISSE	AFNT=	n.	10005	065	
T T	PUT	۶۶	ANCE TOTALE	= 0_3	908F	יוו	MEV/S		
Ť	PHT	22	ANCE PETA	= 0.	1234F	111	VEV/S		1
÷.	DHT	ć¢	ANCE GAMMA	= 0.	1674E	 	MEVIS		
	ر - بیده بنده کنه، شدو بود کنه، نده چنه								
T T	CONTRIBU	TT	ONS A LA PU	TSSAI		51	NUFLEF	TOTALE	(°°)
I									
ī	NUCL IDF	ľ	P.TOTALE	ſ	P.BC	TΛ	T	P.GAMM	4
i T	LΛ140	 T	20.76	 T	3.1	 64	 1	17.12	
Ţ	1132	T	11.20	Ţ	2.	18	T	9.11	1
T	PP144F	ĩ	6.37	Ī	6.1	21	T	0.15	1
Ī	72 95	Ţ	5.89	Ţ	ດູ້.	76	I	5.13	١
T	NB 95F	Ţ	5.63	T	^ .	32	I	5.31	ļ
Ŧ	SP 89	I	4.37	T	4.	7 5	I	0.1	1
7	YOIF	I	4.30	Ţ	4.	27	Ţ	0.03	١
ĩ	BA140	Ţ	3.17	ÿ	1.4	99	Ţ	1.18	۱
I	1123	I	2.78	Ť	1.	11	Ţ	1.67	1
Ţ	CE143	Ţ	2.74	Ţ	1.4	45	T	1.28	۱
T	MA 00	Ţ	2.44	T	3	52	I	0.82	1
Ŧ	NR 97F	ĩ	2.39	Ţ	0.4	7 9	I	1.42	1
۲	PP143	Ţ	2.00	I	2.1	99	Ţ	0.0	1
T	1131	T	1.99	T	0.4	56	T	1.32	ļ
T	7R 97	I	1.84	I	1.3	37	T	0.47	Į
T	CS134F	I	1.71	I	n.]	16	T	1.55	I
J	(F14)	Ť	1.49	τ	1.0	?ר	I	0.47	T
I	YON	I	1.47	I	1.7	٩6	Ţ	0.11	1
Ţ	TF132	I	1.40	Ţ	0.2	25	T	1.15	T
T	NR 97M	Ţ	1.24	T	∩ _ (n i	I	1.24	ו
T	SP 01	Ţ	1.22	T	0.6	50	I	0.63	I
T	XEISSE	T	1.17	I	0.6	55	Ţ	0.52	Ţ
Ĩ	RU103	Y	1.09	Ţ	0.1	13	¥	0,06	Ţ
: · I	PESTE(8)	I	11.18	I	5.7	25	ſ	5.92	1
: •									

TEMPS DE PERDIDISSEMENT= 0.1000F 075 Ŧ T T PUISSANCE TOTALE= 0.1501E 11 MEV/S I T PUTSSANCE BETA = 0.6919E 10 MEV/S- 1 T PUISSANCE GAMMA = 0.8087E 10MEV/S T T I CONTRIBUTIONS & LA PUISSANCE PESIDUELLE TOTALE (") " Ţ -----I NUCLINE I P.TOTALE I P.RETA I P.GAMMA T ----T J T T T Ţ Ŧ Ţ Ŧ T Ţ T ۳ Ŧ 2.4 T PESTE(") I 3.36 3.64 7.01 I T Ţ TEMPS OF REFROIDISSEMENT= 0,1000F 085 Ŧ ĩ т T PUISSANCE TOTALE= 0.4571E LOMEV/S T T PUISSANCE RETA = 0.2627E 10MEV/S Ŧ Ţ PUISSANCE GAMMA = 0.1944E 10MEV/S T - 2 CONTRIBUTIONS A LA PUISSANCE RESIDUELLE TOTALE (") T Ţ Ţ T NUCLIDE I P.TOTALE I P.BETA I P.GAMMA I

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TEMPS DE PEERDIDISSEMENT= 0,1000E 095 I T T 1 PUISSANCE TOTALE= 0.68720 09MEV/S I T PUTSSANCE BETA = 0.3922E 09MEV/S Y Ŧ PUISSANCE CAMMA = 0.2949F 09MEV/S τ T T I CONTRIBUTIONS A LA PUISSANCE RESIDUELLE TOTALE (") T Ţ Ŧ • I NUCLIDE I P.TOTALE I P.BETA I P.GAMMA I

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</ta T F Ţ Т Ŧ Ţ T T Ţ ĩ --------[PESTE(%) I 1.74] 0.91 I 0.83 T ----

TEMPS OF REFERCIDISSEMENT= 0.1000F 105 1 T Ŧ Ŧ PHISSANCE TOTALE= 0.1739E 09MEV/S T Ŧ PUTSSANCE PETA = 0.1168E 09MEV/S Ŧ Ŧ PUISSANCE GAMMA = 0.5711E OBMEV/S CONTRIBUTIONS A LA PUISSANCE RESIDUELLE TOTALE (7) T T . I NICLIDE I P.TOTALE I P.BETA I P.GAMMA I I Y 90F I 48.35 I 48.12 I 0.23 I I RA137M I 32.25 I 0.0 I 32.25 I I CS137 I 9.11 I 9.11 I 0.0 I I SP 90 I 9.07 I 9.07 I 0.0 I -----• _____ ----IRESTE(9) I 1.22 I 0.86 I 0.36 T _____

146

Table A-IV: 235 U thermal fission, case A4

**** 2 -TEMPS DE REFROIDISSEMENT = C.1COCE 015 I T T 1 PUISSANCE TOTALE= 0.3762E 12MEV/S I t PUTSSANCE BETA = 0.1952E 12MEV/S Ŧ T T PUTSSANCE GAMMA = 0.1810F 12MEV/SŢ ____ - • . T T I CONTRIBUTIONS A LA PUISSANCE RESIDUELLE TOTALE (%) I T T - : *-----I NUCLIDE I P.TOTALE I P.BETA I P.GAMMA I

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 Y
 99
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 C.577
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 1.221
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 SR
 95
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 1.221
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 0.76
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 95
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 1.018
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 98
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 1.019
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 0.229
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 SR
 89
 I
 T 0.71 T RB 03 3.4 I I _____ 29.52 I 23.88 I I PESTE(%) I 53.41 I · ****

_____ TEMPS DE REFROIDISSEMENT= 0.1000E 025 T 1 T I PUTSSANCE TOTALE= 0.3010F 12MEV/S Ι T PHISSANCE BETA = 0.1516E 12MEV/S Ţ T PUTSSANCE GAMMA = 0.1494E 12MEV/S I Ŧ ſ I CONTRIBUTIONS A LA PUISSANCE RESIDUELLE TOTALE (%) I I
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 NUCLIDE
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 P.TOTALE
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 CS147
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 2.65
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 134F
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 P0 01
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 P0 07F
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 2.37
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 0.51
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 1347
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 2.37
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 0.15
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 1.49

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 <td T NUCLIDE T P. TOTALE I P. PETA T P. GAMMA I Y RESTE(") I 42.91 I 23.16 I 19.74 I _____

****** TEMPS OF REFROIDISSEMENT= 0.1000E 035 Ţ Ι T I T PUISSANCE TOTALE= 0.1990E 12MEV/S I PUISSANCE BETA = 0.9443E 11MEV/S T Ţ PUISSANCE GAMMA = 0.1046E 12MEV/S Ţ I 2 -- 1 Т T T CONTRIBUTIONS & LA PUISSANCE RESIDUELLE TOTALE (%) I T T
 Image: Second ____ T NUCLIDE I P.TOTALE I P.BETA I P.GAMMA I T RESTE(3) I 30.25 I 15.03 I 15.22 I

TEMPS OF REFROIDISSEMENT= 0.1000E 04S ۲ Ţ T Ī PUISSANCE TOTALE= 0.1179E 12*EV/S T I DISSANCE BETA = 0.5296E 11MEV/S t ſ PHISSANCE GAMMA = 0.6489F 11MEV/S T I 2 -T T T CONTRIBUTIONS A LA PUISSANCE RESIDUELLE TOTALE (3) I T I
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 2. T NUCLIDE I P.TOTALE I P.PETA I P.GAMMA I TRESTE(?) I 22.97 I 11.95 I 11.01 I

****** TEMPS DE REFROIDISSEMENT= 0.1000E 055 I Ţ I T PUISSANCE TOTALE= 0.58785 JIMEV/S I T PHISSANCE BETA = 0.2682E 11MEV/S T T PUTSSANCE GAMMA = 0.2196F 11MEV/S I Ţ · ~ * 2.4 Ŧ . Y CONTRIBUTIONS A LA PUISSANCE RESIDUFULE TOTALE (%) I T τ *-----************

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 P.GAMMA
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 T NUCLIDE I P.TOTALE I P.BETA I P.GAMMA I _____ ------J PESTE(*) I 15.90 I 7.10 I 8.80 I _____ ----

• TEMPS DE REFROIDISSEMENT= 0.1000E 065 I Ţ I T PUTSSANCE TOTALE= 0.2871F 11MEV/S T 1 PUTSSANCE BETA = 0.1214E 11MEV/SI T PUTSSANCE GAMMA = 0.1657E 11MEV/S I Ţ _____ : ĩ T T CONTRIBUTIONS & LA PUISSANCE RESIDUELLE TOTALE (%) I I Ŧ T NUCLIDE I P. TOTALE I P. PETA I P.GAMMA I

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 *********** T SR 91 I 1.12 I C.55 I 0.57 I -----T RESTE(%) I 10.31 I 5.27 I 5.04 I *-----_____ _____

*** *** I TEMPS OF REFPOIDISSEMENT= 0.1000E 075 T T T Ţ PUISSANCE TOTALE= 0.1493E 11MEV/S 1 PHESSANCE BETA = 0.6710E 10MEV/S τ T PUISSANCE GAMMA = 0.82198 LOMEV/S T Ŧ T CONTRIBUTIONS A LA PUISSANCE RESIDUELLE TOTALE (3) I 1 T NUCLIDE I P.TOTALE I P.BETA I P.GAMMA I

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 **** ------ ---------. - 1 I RESTE(9) I 5.85 I 2.47 I 3.38 I 2-**** -----T TEMPS DE REFROIDISSEMENT= 0.1000E C8S I T I PUISSANCE TOTALE= 0.4800E 10MEV/S T I Ţ PHISSANCE BETA = 0.2740E 10 MEV/SŢ PUTSSANCE GAMMA = 0.2060E 10MEV/S T I Ť T T CONTRIBUTIONS A LA PUISSANCE RESIDUELLE TOTALE (3) I T T . ------T NUCLIDE I P.TOTALE I P.EETA I P.GAMMA I

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 CF144
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 I ***** -----...... -----_ _ _ _ _ _ I RESTE(R) I 3.92 I 2.56 I 1.36 I -----

Table A-IV: (cont'd)

2 -TEMPS DE REERDIDISSEMENT= 0.1000E 095 1 1 T T PUISSANCE TOTALE= 0.6953E COMEV/S T Į PUISSANCE BETA = 0.2882E COMEVIS T I Ť PUISSANCE GAMMA = 0.3071E 09MEV/S I • - 2 Ţ T. J CONTRIBUTIONS A LA PUISSANCE RESIDUELLE TOTALE (%) I 7 Ţ 2----I NUCLIDE I P.TOTALE I P.PETA I P.GAMMA I

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 CS134F
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 PR344F
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 1.33</td * -----T PESTE(3) I 1.05 I 1.65 I 0.26 I ****

TEMPS OF REFROIDISSEMENT= 0.1CORE 105 I I 1 I T PUISSANCE TOTALE= 0,1567E COMEV/S 1 PUISSANCE BETA = 0.1031E 09MEV/S Ţ T T PUISSANCE GANMA = 0.5355E CAMEV/S T 2. T T T CONTRIBUTIONS & LA PUISSANCE RESIDUELLE TOTALE (%) I T T - 1 I NUCLIDE I P.TOTALE I P.PETA I P.GAMMA I 2-----

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 -----. ----T RESTE(%) T 1.11 I C.61 I 0.50 I *_____ ____ _____ -----

Table A-V: ²³⁹ Pu fast fission, case A5

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T	ተም	DS	P P	REFE	110	ITSSF	VENT= "	•1000	F ^1 S	
Ţ										
Y	PHI	551	ANCE	: TO1	AL	= 0.	3018F 1	2NEV/	S	
Ŧ	0111	\$\$/	NCE	8.61	Δ.	= 0.	1559F]	2NEV/	S	
T	PHT	551	ANC P	GAN	MA	= 0.	14598 1	2NEV/	S	
T										
T CONT	FR TBU	TT	INS	AL	PI.	ITSSAL	VCE RES	IDLFL	LE TOTALE	(8)
ſ										
						·			*******	
NUCL	INE	Ţ	Р.	TOT	LE	T	P.BET	Ά	I P.GAMM	Δ.
	36F	I		`` `	9	Ţ	1. ^	6	I 1.33	
112	84F	T		?,?	1	Т	C. 5	1	I 1.80	
C 5 1 4	12	Ţ		2.1	1	I	1.1	^	I 1.00	
Y Ç	76	Ţ		1.7	3	Ţ	r.9	a 1	I 0.94	
MOIN	13	Ţ		1,7	7	τ	r. 4	7	I 121	
7010	14	J].7	7	Ţ	0.7	9	1 0.97	
(dis	IPF	I		1.6	6	T	s	6 1	1.10	
1114	in -	Ϋ́Ι		1.4	6	Ţ	5.2	9	I].37	
1 5 1 4	. ?	Ť		1.6	3	T	2.4	4	1.18	
רוחד	25	Ţ		1.5	1	Ţ	1.0	1 1	r 0,59	
(513	0	Ţ		1.6	ā -	Ţ	1.1	3 1	1 0.47	
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7136F	T	2.66	I	1.13	I	1.48
CS140	I	2.32	I	1.22	I	1.11
¥ 96	Ţ	2.20	I	1.13	I	1.67
10104	Ţ	2.19	I	C. 94	I	1.15
rs138F	T	1,97	1	Č. 67	T	1.30
L1147	I	1,07	Ţ	0.34	I	1.62
1 1142	T	1.03	Ţ	C. 53	Ţ	1.47
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1137	ו ד	1.23	L T	° • 7 7	T T	0.78
Y 94	Ť	1_29	Ť	r_ 98	Ť	0,30
Yor	ī	1,73	Ť	6,96	Ť	0,27
(n ) F	ť	1.18	ī	0.43	ŗ	7,75
PB 91	Ť	1,18	Ī	r . 85	ŗ	1.33
50 03	T	1.18	τ.	6.53	Ĩ	0.64
85130	I	1.17	I	r_ 89	T	n.28
PA141	T	1-15	I	1,74	T	0.41
TE133E	T	1.14	I	C _ 44	T	0,70
14144	1	1.08	Ţ	C.71	I	0.37
NA GAE	T	1,07	Ţ	], ]]	I	ຳ5
MO105	T	1.04	I	<b>^.</b> 32	I	n.71
TE136	I	1.13	T	<b>°.</b> 34	T	0.69
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Ţ	11142	Ţ		2.3	n -	٤		٢.,	77	Ţ	2.13
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Ŧ	PA141	Ŧ		1.5		r		1	13	Ţ	0.56
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Ţ	1132	r	8,79	I	1.70	I	7.10
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Ţ	[132	T	4.04	T	2.61	T	2.43
Ţ	NA 97E	T	3.51	I	1.43	I	2.08
۲	1134F	J	3.47	ĩ	r.76	I	2.71
I	PHIDEE	Ŧ	3.45	Ŧ	3.03	J	0.42
T	Y 02	T	2.94	Ι	2.43	I	0.41
۲	10342	I	2.92	Ţ	C.77	Ţ	2.15
Ţ	70 97	Ţ	2.73	I	2.03	ĩ	0.70
T	X=135F	I	2.59	Ţ	1.39	I	1.22
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Ŧ	7P 95	I	2,44	I	C. 32	I	2.13
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RH106F	I	6.70	I	5.99	I	0.82
70 05	Ţ	4.71	Ι	6.61	Ţ	4.10
PR144F	Ţ	4.56	I	4.45	I	0.11
F1]] 13	T	4.53	T	0.53	I	4.31
NA OFF	T	4.50	T	C.26	I	4.24
1133	1	3.45	I	1.38	ĩ	2.08
PA147	I	2.05	I	1.85	T	1.1.)
VC 90	I	2,95	I	1.06	τ	n <b>,</b> 99
1131	Ţ	2,57	I	C.85	I	1.72
NR 97F	I	2,47	I	1.00	Ţ	1.46
CE143	J	2.14	Ĩ	1.14	I	1.00
7P 97	Ţ	1.90	I	1,41	I	0.49
Y 91F	J	1.78	ĩ	1.77	I	0.01
TF132	I	1.71	I	C.31	T	1.47
CF141	T	1.67	J	1.14	I	0.53
PRI43	I	1.64	τ	1,64	ĩ	n.0
YE135E	I	1.53	I	C.81	I	0.72
XF133F	I	1.46	I	C.81	Ţ	1,65
<u>NB 03N</u>	Ι	1.28	I	0.0	I	1.28
28 8 <b>9</b>	T	1.14	I	1.14	I	n.0
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----TEMPS DE REFROIDISSEMENT= 1.1000E 075 T 1 T T PHILSSANCE TOTALE= 0.1250E 11MEV/S T I PHISSANCE BETA = 0.5492E 10YEV/S T I PHILSSANCE GAMMA = 0.7005F LOMEN/S T I -----÷ -Ţ τ. T CONTRIBUTIONS A LA PUISSANCE RESIDUELLE TOTALE (%) I T Ŧ · · · · · T NICLIDE I P. TOTALE I P. PETA T P.GAMMA I _____ :-----2.15 2.72 1.89 1.89 1.89 1.2.10 1.2.25 1 0.37 1.44 1.1 2.15 2.05 T CD 99 I I I I T NO147 I 1.02 J C.53 I 0.44 I ---------_____ - * _ _ _ _ _ _ _ T RESTR(7) T 7.79 T 2.08 T 4.71 I TEMPS OF REFERDIDISSEMENT= 0.1000E CAS T T Ŧ I PUTSSANCE TOTALE= 0.4000E 10MEV/S Ţ I PUISSANCE BETA = 0.2550E 10MEV/S Ţ T PUISSANCE GAMMA = 0.1450F LOMEV/S T . - * 1-T I CONTRIBUTIONS A LA PUISSANCE RESTOUELLE TOTALE (3) I T T •-----T NUCLIDE T P. TOTALE I P. PETA I P.GAMMA I ----- 

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Review Paper No. 5

# FISSION PRODUCT NUCLEAR DATA REQUIREMENTS FOR THE DETERMINATION OF NUCLEAR FUEL BURNUP: A REVIEW

W. J. Maeck

Allied Chemical Corporation* Idaho Chemical Programs* Idaho Falls, Idaho USA

ABSTRACT

This paper reviews and expresses specific needs for improved fission product nuclear data for the determination of nuclear fuel burnup. The various techniques for measuring burnup, both destructive and nondestructive, are discussed with regard to applicability and to fission product nuclear data requirements. The highest priority fission product nuclear data requirements are for fast reactor fission yields as a function of neutron energy and for decay scheme data for radionuclides used as burnup monitors. New data showing the variation of fission yields with neutron energy and the need to associate all fast fission yields with a well defined spectral index are given.

### 1. INTRODUCTION

The economical production of electrical power based on nuclear reactors requires 1) that the fuel be operated at both a high temperature and high power density to maximize the integrated power output, and 2) a viable fuel reprocessing and fabrication cycle. In the development of nuclear fuels and in the operation of nuclear reactors, it is imperative that there be accurate methods for the evaluation of the fuel and reactor performance. An important criterion in determining fuel performance is an accurate determination of the total fissions and the fission rate. This is accomplished by a burnup determination. Errors in a burnup measurement introduce errors in fuel design and operation, nuclear physics calculations, shielding requirements, design of transportation equipment, and fuel reprocessing equipment, all of which affect power generating costs.

The term burnup is used to express the degree of fuel consumption by the fission process relative to some property of the fuel prior to irradiation. It is defined several ways. In the nuclear power industry and to some extent in irradiated fuel experimental programs, burnup is defined as megawatt-days of thermal energy produced per metric ton of heavy atoms initially present in the fuel. Another definition for experimental fuels is the number of fissions per cubic centimeter of the fuel.

In this review, burnup will always mean:

Burnup = Atom percent fission =  $\frac{\text{number of fissions x 100}}{\text{initial number of total heavy atoms}}$ 

This definition is more fundamental in that it relates directly the number of events of interest, fissions, to the quantity of most interest, the number of fissile and fertile atoms, in the fuel at the beginning of the irradiation. In other definitions, it is necessary to have knowledge of either a physical constant such as energy of fission, or a property of the fuel such as bulk density, to calculate burnup from measurements provided by a chemist. Although the errors in these conversion factors are small, their use introduces an additional uncertainty in the burnup value obtained.

Within the megawatt-day/ton (MWd/T) and atom percent fission definitions of burnup, there are variations. These variations have, in the past, resulted in invalid comparisons being made of the performances of irradiated fuels. The megawatt-days of energy produced by a fuel is of prime concern to the power reactor operator while the megawatt-days of energy deposited in a fuel is of prime concern for an evaluation of its irradiation performance and stability. The energy produced by a fuel will, in a fast reactor such as EBR-II, be 5 to 10% higher than the energy deposited. In both the MWd/T and atom percent fission definitions, the quantity against which the energy or number of fissions is referenced is not always defined the same way. The "ton" in the MWd/T definition has meant, in addition to metric ton of heavy atoms, metric ton of fuel (U + Pu + O or C) and short ton of heavy atoms. In the atom percent fission definition, only the number of fissile atoms rather than the total number of heavy atoms is sometimes used as the reference quantity. In a low enrichment ²³⁵U fuel, the difference between the per fissile atoms and the per total heavy atoms burnups is very large; for high enrichment fuels, the difference is less than 10% (relative).

From burnup measurements, both the total number of fissions and the fission rates are calculable. The total number of fissions is a measure of the thermal energy produced by a fuel. This thermal energy is required to establish fuel warranties (some cores will cost on the order of \$100,000,000) and to evaluate the performance of experimental fuels. Fission rates are used to evaluate such fuel performance characteristics as power levels, fuel and cladding temperatures, and fuelcladding gap conductances. Fission rates measured by foil activations also are used to establish flux distributions in critical assemblies and in test reactors.

The primary function of this paper is to review the current status of burnup measurement technology with regard to the adequacy of the nuclear data requirements for the various techniques and to point out where improved nuclear data, especially for the fission products, are required to improve the reliability of the various techniques. A secondary function will be to discuss the chemistry problems associated with a burnup measurement and to relate both the nuclear data and chemical analysis uncertainties to the total error.

### 2. TECHNIQUES FOR DETERMINATION OF BURNUP AND APPLICABILITY

Absolute burnup values are determined most accurately by destructive chemical analyses; relative burnup is most rapidly determined by nondestructive analyses. The former is discussed below and the latter in Section 2.2.

### 2.1. Destructive analysis

Four destructive chemical techniques have been used for determining burnup; (1) measuring a fission product monitor and the residual heavy atom contents of a dissolved fuel specimen and calculating the burnup from these values and the yield of the fission product, (2) measuring the plutonium and uranium isotopic ratios on dissolved specimens of both the irradiated and unirradiated fuel and calculating the burnup from these values and nuclear cross section data, (3) measuring the heavy atom contents of dissolved specimens of both irradiated and unirradiated fuel and calculating the burnup from the change in these values, and (4) correlating the isotopic composition of selected fission products such as  83 Kr/ 84 Kr,  134 Cs/ 137 Cs or heavy elements with atom percent fission.

Each of these techniques has particular advantages and disad-vantages.

The major emphasis in the discussion of destructive burnup techniques and the nuclear data requirements that follow is devoted to the fission product-residual heavy atom technique because it has the widest range of applicability and is the only one that can provide high accuracy for fast reactor fuels.

2.1.1. Fission product monitor-residual heavy atom technique

The irradiated fuel specimen is dissolved and the fission product monitor and heavy atoms are determined. The computational relationship is

Burnup = a/o F = 100 
$$\frac{A/Y}{H + A/Y}$$

where

a/o F = atom percent fission

- A = determined number of atoms of fission product monitor
- Y = effective fractional fission yield value of A
- H = determined number of residual heavy atoms.

The successful application of this technique requires accurate measurements of the fission product monitor and heavy atoms and an accurate value for the effective fission yield. When using this technique for determining burnup, it should be recognized that the fission yield error is constant while the measurement error can be both random and systematic.

When isotope dilution mass spectrometry is used as the measurement technique for both the fission product and the heavy elements, the measurement errors are established by repeated analyses of synthesized solutions of uranium, plutonium, and the fission product elements. These solutions are prepared from certified uranium and plutonium compounds and/or metals and highly pure and stoichiometric fission product element compounds. If biases are detected in the analyses of these solutions, modifications are made in the methods to eliminate them. Hence, the use of this technique should minimize the systematic error in the measurement with the result that the major error is the random component. Currently, the isotope dilution mass spectrometric technique is capable of 0.25 relative percent uncertainty in the measurement of fission product and heavy atoms.

For a radiochemical analysis of the fission products and/or heavy elements in the fuel sample, significant systematic errors can exist because of errors in the decay schemes,  $\gamma$ -ray intensity values, halflives, detector calibrations, and data reduction (baseline subtractions in  $\gamma$ -ray spectra). Thus, the systematic errors in the nuclear data and the data reduction can be more than the random error of the measurement.

The effects of the measurement errors and uncertainties in the fission yield values on the total burnup error were studied and are shown in Figure 1. Increasing error values were placed on each variable of the basic burnup relationship discussed above and propagated to give the overall error in the burnup. For this study, the U/Pu ratio of the heavy element component was fixed at 3. Figure 1 shows the magnitude of the error in the burnup determination as a function of the measurement error for fission yield uncertainties of 1, 2, 3, and 4%. At anticipated burnup, a 4% uncertainty in the yield data will result in an error of 4.1, 4.2, and 6%, respectively, when the measurement error is 0.5%, 1.0%, and 3.0%. At a 1% uncertainty in the yield data, the burnup errors would be 1.2, 1.5, and 4.3%, respectively, for the same measurement errors. For this last case, if one assumes that the major fraction of the measurement error in a radiochemical analysis is the uncertainty in the nuclear data, elimination of this error would significantly reduce the overall burnup error. Thus, the needs for accurate yield data and nuclear decay data are clearly demonstrated.

For a determination of burnup based on isotope dilution mass spectrometric measurements, the dominating error is the uncertainty in the fission yield, even if this value is known to 1%. For a radiochemical analysis, the situation is reversed with the source of major error being the uncertainty of the nuclear decay data and the detector calibration, especially when the fission yield is well known.

Another point to be considered is the interplay of the random and systematic errors in the measurements and the constant error of the fission yield when the objective is to compare a series of fuel specimens of the same composition irradiated under similar conditions to determine the relative burnup. The error in the relative burnup values is due solely to the random errors of the measurements of uranium, plutonium, and the fission product monitor. For this case, which can be the case for non-destructive assay, the line that applies in Figure 1 is the one for zero uncertainty in the fission yield value.

The above discussion has assumed no error in sampling; however, sampling uncertainties can significantly contribute to the total error (ie, if the operator does not achieve total dissolution of the sample or if there has been migration or fractionation of the fission products or heavy elements in the sample being analyzed).





2.1.2. Heavy atom ratio technique

The change in the heavy element isotopic composition of a nuclear fuel can be used to determine the atom percent fission. This method is applicable to thermal reactor uranium and plutonium fuels but does not apply to fuels containing thorium or  233 U before irradiation. Its application to fast reactor fuels is doubtful because of the multiplicity of nuclear reactions which occur and the uncertainty in the nuclear constants which are required in the calculation.

There are several computational relationships which are used to measure burnup from changes in the isotopic composition of the heavy elements. One basic relationship is:

$$F_5 = N_8^\circ [(R_{5/8} - R_{5/8}) - (R_{6/8} - R_{6/8})]$$

where,

 $F_5 = \text{atom percent fission from } ^{235}\text{U}$   $N_8^\circ = \text{atom percent } ^{238}\text{U} \text{ in the preirradiated fuel}$   $R_5^\circ/_8, R_6^\circ/_8 = \text{atom ratio } ^{235}\text{U to } ^{238}\text{U, and } ^{236}\text{U to } ^{238}\text{U in the preirradiated fuel}$   $R_5/_8, R_6/_8 = \text{atom ratio } ^{235}\text{U to } ^{238}\text{U, and } ^{236}\text{U to } ^{238}\text{U in the final irradiated sample.}$ 

This relationship is applicable when more than 10 percent of the  235 U is consumed. It is assumed that no loss of  238 U or  236 U occurs during the irradiation. This relationship is not recommended for low burnup or where recycled uranium is used in the fuel and  $R_{6/8}^{6}$  ratio is large with respect to  $R_{6/8}$ .

For the non-ideal cases, other relationships can be used [2]; however, these require a knowledge of  $\alpha_5$ , the capture-to-fission ratio for ²³⁵U, which may be in error by as much as 20%. This error is not from a lack of knowledge of the variation of  $\alpha_5$  with differential neutron energy data but rather results from the inability of the experimenter to define the best integral value for the particular reactor core and location within the core. For ²³⁵U the capture-to-fission ratio can vary from about 0.17 for well moderated thermal reactors to as high as 0.65 for intermediate-neutron-spectrum reactors [3] and as low as 0.1 [3] for fast reactors.

Other relationships have been developed to calculate the atom percent fission due to  239 Pu and  241 Pu; however, these require a know-ledge of a for  239 Pu and  241 Pu [2].

Another prevalent source of error in the heavy atom ratio technique for the determination of burnup and one over which the chemist doing the analysis has no control is the equivalency of the pre- and postirradiated fuel specimens analyzed. It has been the observation of many that the equivalency requirement of the pre- and postirradiated fuel samples has not been met on a number of occasions, despite claims to the contrary by the individuals submitting the samples for analyses. For example, cases have been observed where the uranium and plutonium analyses (chemical and/or isotopic) of so-called duplicate specimens of the preirradiated fuel do not agree while replicate analyses of the same sample do. This situation is likely for fuels prepared by blending uranium and plutonium oxides, particularly when different batches of the oxides are used to manufacture a fuel lot.

As more accurate cross section values are experimentally determined, the accuracy of burnups calculated by this technique will improve. However, it is doubtful that accuracies comparable to those obtained by the fission product monitor-residual heavy atom technique can be achieved, even at high burnup. It is important to note that the fission product monitor technique is one of the key techniques used to determine burnup and it will always be the reference method against which the heavy atom ratio technique must be evaluated.

A more detailed description of these calculational techniques and specific applications is given in Reference [2] for thermal reactors and Reference [1] for fast reactors.

2.1.3. Heavy atom difference technique

In this approach, the number of fissions is established by subtracting the number of heavy atoms measured in the irradiated fuel specimen from the number of heavy atoms measured in an equivalent specimen of preirradiated fuel. In principle, this is the best method for determining burnup because fission yield and capture-to-fission ratio data are not involved in the calculation. It is especially suited to high enrichment fuels which have had high burnup and to specially prepared irradiation capsules which are used to accurately determine specific nuclear data.

The applicability of this method to the determination of burnup on light water power reactor fuels and fast breeder reactor fuels is highly questionable for two reasons. First, the number of fissions relative to the number of total heavy atoms is relatively small, usually less than 10%. If the number of uranium and plutonium atoms is determined with a relative standard deviation of 0.25%, (this precision is routinely obtained in experienced laboratories using isotope dilution mass spectrometric techniques) the propagated error in the measured number of fissions is 18% for 2% burnup and 3.4% for 10% burnup.

The second reason is the practical difficulty of obtaining preand postirradiated fuel specimens that are matched on an equivalent weight basis. At high burnups, fuel melting and swelling, heavy atom migration, and cladding-fuel reactions occur. The assumption, that a weighed specimen of the irradiated fuel contains all the heavy atoms initially present less those that have fissioned and the same ratio of fuel to cladding as the preirradiated specimen, is untenable.

Specific nuclear data requirements for this method are a knowledge of the capture cross section for some of the minor heavy isotopes and half-life data for some of the shorter lived heavy elements. In particular, in the irradiation of  235 U to high burnup, corrections for the capture of  236 U to  237 U and its subsequent decay to non-uranium isotopes must be made. Long term irradiations of Pu require reliable half-life data for corrections to the  241 Pu decay. Generally, these corrections are small and only required in the most exacting work.

### 2.1.4. Isotope correlations

At the current stage of development, this is a semi-empirical method which is based on the relationship of certain isotope ratios to burnup. Generally, the fission product isotope ratios which change with burnup are those in which one of the isotopes, A, has a large capture cross section for the formation of the A + 1 isotope. In this case, the change in the A/A+1 ratio is directly related to the neutron flux and, hence, to burnup. Early conceptual studies of this approach to measuring burnup were reported [4] in 1965. More recently Koch [5,6,7] has reported experimental data showing many different correlations of isotope ratios with burnup. Some of the more significant fission product ratio correlations are: 1) the ratio of 84Kr/83Kr which shows a linear increase with burnup, 2) the ratio of 132 Xe/131 Xewhich also shows a linear relationship with burnup, and with ²⁴¹Pu and  242 Pu production, and 3) the ratio of  136 Xe/ 134 Xe which correlate with ²³⁹Pu buildup. Robin and co-workers [8] and Eder and Lammer [9] and Natsume and co-workers [10] have recently suggested that the ratio of ¹³⁴Cs/¹³⁷Cs can be used as a measure of fluence and, hence, possibly The majority of the isotope correlation studies have been for burnup. thermal fuels.

Other fission product correlations with burnup involving nuclides of different elements which have been studied are  $^{154}Eu/^{144}Pr$ ,  $^{134}Cs/^{144}Pr$ , and  $^{154}Eu/^{137}Cs$  [10].

Recent data by Umezawa and co-workers [11] show some interesting correlation of transplutonium nuclides with burnup in JPDR-1 spent fuel. They report correlated relationships of  241 Am,  242 Cm and  244 Cm to  238 U and  242 Cm, and  244 Cm to  242 Am. In addition they show that the slope of heavy nuclide ratios to burnup divided by the mass difference between the two nuclides is essentially 1 for all relationships. That this relationship is the same for all initial fuel loading remains to be proven.

To improve the calculations which are required to substantiate the correlation of these measured ratios to burnup requires a significant improvement in the neutron capture cross section data for the specific fission product nuclides and heavy element nuclides being used in the correlation study.

At this time, it appears that the primary usefulness of this technique is one of a supplementary method to the more conventional techniques and is not being suggested as a standard method for measuring burnup.

#### 2.2. Non-destructive analysis

Several methods have been used or proposed for the non-destructive determination of burnup on irradiated fuel samples. In general, these methods involve the use of specific nuclear properties ( $\gamma$ -ray emission, delayed neutrons, neutron resonance adsorption, and decay heat) of selected fission products or residual heavy elements. The principal merit of a non-destructive analysis is the routine and rapid determination of relative burnup.

## 2.2.1. Gamma-ray scanning

This technique has been widely used for several years in many laboratories throughout the world for the non-destructive determination of both the relative and absolute burnup. Significant improvement in the measurement of specific  $\gamma$ -rays has resulted from the use of Ge(Li) detectors; however, uncertainties in the nuclear data still remain, and has introduced significant errors in the final results. The successful application of this technique to the absolute measurement of burnup requires knowledge of the following:

- a) Half-life of the fission product being measured
- b) Irradiation history and flux levels for proper in-pile decay corrections
- c) y-ray transmission characteristics of the fuel
- d) Detector efficiency
- e) y-ray branching and intensity factors
- f) Fission yield of nuclide being measured

Eder and Lammer [9] and Hick and Lammer [12] recently reviewed the uncertainties in fission product nuclear data with respect to the  $\gamma$ -ray spectrometric analysis on spent fuel and the reader is referred to these reports for detailed discussions.

In addition to the above listed items, other problems characteristic of the fuel or the reactor should be considered, especially if absolute integrated burnup is desired and there are multiple sources of fission. Natsume and co-workers [10] have shown that for a BWR-type reactor, such items as the steam void distribution, control rod pattern, and irradiation history can affect the results obtained in the complete  $\gamma$ -scanning of a fuel rod. Migration of certain fission products, especially ¹³⁷Cs, has long been recognized. The redistribution of uranium and plutonium both axially and radially in oxide fuels, has been reported [15,16] and in some cases, has been observed to be as high as 50% [17].

To attempt to use this technique for the measurement of absolute burnup, requires an extensive calibration program involving many destructive analyses. The most useful application of this technique is to establish relative burnup where systematic errors in the nuclear data cancel and the majority of the error is due to the randomness of the measurement.

### 2.2.2. Activation analysis

This technique is primarily used to measure the heavy atom content of the fuel. Its primary application has been the non-destructive measurement of the fissionable material content of unirradiated fuel. Several nuclear reactions have been proposed or used. Included are 1) measurement of selected short-lived radionuclides, 2) measurement of delayed neutrons, and 3) measurement of prompt gamma rays, all after exposure to a beam of neutrons. One involved technique based on first measuring the ratio of 137Cs to 106Ru and then reirradiating the fuel for 1 day and measuring the 140Ba/140La to determine the ratio of U to Pu is being studied [18].

The successful application of any form of activation analysis to the non-destructive analysis of irradiated fuel samples also requires an extensive calibration program involving destructive analyses. In addition, each method requires extensive knowledge of many nuclear constants both for the fission products and heavy elements, such as half-lives, decay schemes including prompt  $\gamma$  and delayed neutrons. fission yields,  $\gamma$ -ray adsorption, and heavy element cross sections.

As a non-destructive method for burnup based on the loss of heavy

atoms, this technique may be applicable to highly enriched fuels (MTRtype) having high burnup; however, for low enrichment power reactor fuels or fast breeder fuels, this technique does not appear promising because the fuel composition is complex and the loss of heavy atoms is low (see Section 2.1.3).

# 2.2.3. Neutron adsorption - transmission

Another technique for the non-destructive determination of the residual heavy atom content of irradiated fuel is being studied by Priesmeyer [19]. This method for  235 U is based on the neutron resonance adsorption at 8.78 eV, 11.67 eV, and 12.4 eV and requires the use of a fast-chopper spectrometer. Initial studies indicate that this method is applicable to highly irradiated material because there are no strong fission product resonances at these energies and the detector system is not sensitive to high level gamma radiation. Analysis of unirradiated and irradiated fuels shows a 5% agreement with destructive analysis.

The applicability of this method to determining burnup by the loss of  235 U may be greater than activation analysis but problems related to plutonium adsorption must be evaluated. Currently, available resonance adsorption cross sections for the fission products and heavy elements needs to be examined.

# 2.2.4. Calorimetry

The calorimetric determination of decay heat as a possible nondestructive measure of burnup is being examined by Devillers and cocorkers [20] and Debertin and Ramthun [21,22]. The principle of this method consists of correlating burnup with the post-irradiation measurement of the heat power originating from the radionuclides formed in the fission process.

The instrument constructed by Debertin [21,22] is a small adiabatic device and has been used to measure small sections of highly enriched  235 U fuel plates and specially prepared samples. Initial studies with these well controlled samples indicate that burnup results comparable to those obtained by  $\gamma$ -ray spectrometry are possible. Construction of a device to measure entire MTR-type elements is in progress. Similar results have been reported by Devillers [20] for the measurement of small samples irradiated in a fast reactor.

The fission product nuclear data requirements for this method vary because different radionuclides provide the major fraction of the heat output for different degrees of burnup, irradiation times, and cooling times. Certainly fission yields,  $\gamma$  and  $\beta^-$  decay energies and heat capacities are important considerations.

This approach shows promise as a possible non-destructive method for burnup based on initial well controlled experiments; but, its applicability to power reactor fuels which have had long reactor residence times and have been exposed to different fluxes in the fuel management cycle, will require extensive work.

### 3. FISSION PRODUCT AND HEAVY ELEMENT NUCLEAR DATA REQUIREMENTS

### 3.1 Fission yields

Without a question, the fission yield is the most important fission product nuclear data parameter required for the accurate determination of nuclear fuel burnup by any of several available methods, either destructive and non-destructive. It is most critical to the fission product monitor - heavy atom technique which is emerging as the standard method and is the one to which all other methods are compared. The future needs for more accurate yield data are variable and depend upon the composition of the fuel and whether the data are to be applied to thermal reactor fuels or fast reactor fuels.

At the present time, the most critical need is for fission yield data for fast reactors as a function of neutron energy. As will be discussed later, attempting to generate a single set of fast reactor fission yields is unsatisfactory because the yields are continually changing with energy. Hence, any new data, proposed experiments, or compilations of data must have associated with it, well defined spectral data.

#### 3.1.1. Thermal fission yields

Several compilations [23,24,25,26,27] of thermal fission yield data are available in the literature. Generally, these compilations are in agreement; however, a few differences still exist for a limited number of nuclides.

With respect to burnup measurements, the general conclusion from the majority of experts in the field is that the existing thermal fission yields are adequate and that any new extensive program to again measure thermal yields is probably not economically justifiable. This conclusion applies especially to thermal yields for  235 U and  239 Pu where the uncertainties are estimated to be 1-2%. For  233 U and  241 Pu, the uncertainty levels are not quite as good, 2-3%, but adequate.

However, specific requests for improved thermal yield data still exist. These are: 1)  106 Ru, for  235 U and  239 Pu because of its potential use in estimating the fractional source of fission, 2)  141 Ce for  235 U and  239 Pu, 3)  147 Nd for  239 Pu and  241 Pu, 4)  153 Sm for  235 U,  239 Pu, and  241 Pu, 5) independent yields of shorter lived nuclides which are used in fission rate measurements, 6) yields for some of the shielded nuclides such as  134 Cs,  142 Pr, and  154 Eu, and 7) the 140 mass chain because of some discrepancies noted in the  140 Ba-La and  140 Ce yields. These are isotopes which are used to measure burnup by radiochemical methods. Also, their end member of the mass chain is not easily measured by isotope dilution mass spectrometry and hence, the yields have larger errors.

#### 3.1.2. Fast fission yields

This is the area in which lie the greatest uncertainties and for which there is the greatest need for improved data. Because of the multiple sources of fission in a fast breeder reactor fuel, the fission yields must be known for a variety of heavy atom nuclides; however, the accuracy requirements vary with the fractional number of fissions. To illustrate this, the sources of fission in two USA fast breeder reactors have been calculated and the required accuracies of the fission yields necessary to obtain an effective fission yield accurate to 1% for each reactor were determined. These calculations were based on the assumption that at some point on the mass yield curve that one nuclide or group of nuclides would have the same fission yield for the two major tissioning nuclides. The results are given in Table I. For these reactors, the fractional sum of the two major sources of fission is essentially constant for burnups ranging from 1-10 atom percent. Similar calculations can be made for other fuel loadings and reactors.

	TABLE I AN	. REQUIRED AC OVERALL 1% AC	CURACIES OF FIS CURACY IN THE E	SION YIELD VAN FFECTIVE FISS	LUES TO OBTAIN ION YIELDS			
		FI	rTF[a]	1000 MWe FBR[b]				
Fissioning <u>Nuclide</u>		Source of Fissions, %	Fission Yield Accuracy, %	Source of Fissions, %	Fission Yield Accuracy, %			
233U								
235U		2	10					
238U		8	5	3	10			
238p	u			1	30			
239 _{P1}	u	85	1	74	1			
240 _{P1}	u	2	10	4	7			
241Pt	u	3	10	17	2			
242P	u			1	30			
[a]	Based 2.9% ²	on a fuel comp ⁴⁰ Pu, 0.4% ^{24]}	position of 0.5% Pu, and <0.1% ²	2 ³⁵ U, 74.4% ⁴² Pu.	²³⁸ U, 21.7% ²³⁹ Pu,			
[b]	Based 5.2% ²	on a fuel comp ⁴⁰ Pu, 2.6% ^{24]}	position of 78.0 Pu, and 0.9% ²⁴	0% ²³⁸ U, 0.3% [™] ²² Pu.	²³⁸ Pu, 13.0% ²³⁹ Pu,			

For  235 U and  239 Pu, Lisman and co-workers [23] have reported yield data with uncertainties of 2-3% for the major fraction of the mass yield curve for fuel irradiated in EBR-I core center. Because of the interest in the yields of the neodymium isotopes for use as burnup monitors, more information exists for these isotopes than any others. Sinclair and Davis [28] have reported neodymium yields with uncertainties of 3-4% for  235 U and  239 Pu for samples irradiated in DFR. Crouch [29] has also reported neodymium data relative to  143 Nd for DFR, but apparently from samples irradiated in a different location in reactor and with no error limits. Robin and co-workers [30] have reported  148 Nd yields for  235 U,  238 U, and  239 Pu and will report updated values and new measurement for  241 Pu and  240 Pu at this conference, all for samples irradiated in the French fast reactor RAPSODIE. Also contributed to this conference (Paper 11b) is a new set of  235 U fast fission yields for samples irradiated in EBR-II [31].

In general, for the accurate determination of burnup for fast reactor fuels, the yield data for ²³⁵U and ²³⁹Pu are inadequate, for ²³⁸U quite unreliable, and for ²⁴⁰Pu and ²⁴¹Pu nonexistent. In addition, no yield correlations with fast reactor neutron spectra have been developed. Table II summarizes the current status of fast fission yield data as viewed by the users of these data for determining burnup and
lists their estimated needs based on various proposed fuel loadings and fuel development studies being conducted in different laboratories.

TABLE II.	FAST FISSION Y	TELDS:
PRESENT	DATA AND NEED	)S
	Relative	%
Heavy Nuclide	Present Data	Need
233 _U	NA[a]	1
235 _U	₂₋₃ [b]	1
²³⁸ U	∿15	5
237 _{Np}	NA	10-30
238pu	NA	10-30
239 _{Pu}	2-3[b]	1
240Pu	NA	5
²⁴¹ Pu	NA	2
242pu	NA	10-30
241 _{Am}	NA	10-30
243 _{Am}	NA	10-30

For a general discussion of fast fission yield measurements and various measurement programs in progress, the reader is referred to Paper 11b of this conference.

To provide that information required for the accurate determination of fast reactor fuel burnup, extensive fission yield measurement programs are under way in the UK by Sinclair [32] using DFR as the irradiation source, by Koch [33] and Robin and co-workers [34] using RAPSODIE, and by Maeck and coworkers in the USA using EBR-II [1].

[a] Reliable data not available.

[b] For EBR-1.

To date, the major request for improved data by those measuring burnup has been for a single set of 1-2% accurate "fast reactor fission yields". Hence, the users, measurers, and compilers of fast yields have tended to treat the fast data much like thermal data. That is, several pieces of independent data are compared to each other to obtain a weighted average fast reactor fission yield. Based on very recent studies in our laboratory in the USA [35], we now believe that this approach is not valid for the evaluation of fast yields, and that a comparison of DFK, RAPSODIE, EBR-I, and EBR-II yield data cannot be made unless the neutron energy spectrum in which the samples or fuel were irradiated is well defined.

To support this conclusion, the change in the isotopic ratio of 143Nd to 150Nd for 235U fast fission as a function of neutron energy for several different sets of data is shown in Figure 2. In this study only the isotopic ratios of neodymium were examined because the relative isotopic composition is easily measured and the absolute reported fission yield values may be biased by systematic errors in the number of fissions used to arrive at the yields. Thus, by only comparing isotopic data or the ratios of the Nd yields, these systematic errors cancel.

The establishment of a neutron energy index was difficult. Several definitions of reactor neutron spectrum have been used; mean neutron energy, median neutron energy, mean and median neutron energy for fission of a given heavy isotope, fraction of neutrons in a given energy range, and the ratio of fission cross sections. We selected the ratio of the fission cross section of  238 U to  235 U ( $\sigma_{f_8}/\sigma_{f_8}$ ) because more data existed for this definition, the neutron energy index could be treated in a linear manuer, and it is a very sensitive index.



Fig. 2 Variations in the ratio of  143 Nd/ 150 Nd with neutron energy.

Fortunately, all of the samples irradiated in EBR-II can be associated with this spectral index because the reactor has been mapped with these monitors or they were included in the sample assemblies irradiated for fission yield measurements [1]. The relative Nd isotopic data reported by Robin and co-workers [30] is believed to have been obtained from samples irradiated in OSIRIS for which a  $\sigma_{fg}/\sigma_{fg}$  value of 0.89 is reported [30]. The Nd data for DFR [28] has not been directly associated with a spectral index but the value is believed to be in the range of 0.035 to 0.04 based on a fission cross section for  238 U in DFR at that time of  $\sim$ 0.04 [36]. An uncertainty of ±10% has been placed on all of the spectral index values.

Of the various Nd isotopic ratios that can be calculated,  143 Nd/ 150 Nd shows the greatest change, 20%, in going from an EBR-I to a thermal spectrum. In this case, the  150 Nd yield is increasing with neutron energy while the  143 Nd yield is decreasing. When other Nd ratios are plotted in the same manner, similar curves with the same energy dependency are obtained but the change is less. From an examination of the changes in the relative isotopic data and an evaluation of reported absolute yields, it appears that the  235 U fission yields for  145 Nd and  146 Nd change little with neutron energy.

Using this observation, this study was carried one step further. By assuming the yields of  $1^{45}$ Nd- $1^{46}$ Nd to be constant with neutron energy, the relative isotopic data and the reported absolute yield data were normalized to  $1^{45}$ Nd and  $1^{46}$ Nd and the percentage change in the fast yields relative to the thermal yields for each isotope was calculated as a function of neutron energy. These data showing the changes in the yields as a function of energy and of mass number are given in Figure 3. None of the data shown in Figures 2 and 3 has been corrected for fission product burnin or burnout by neutron capture. However, this effect, at least for these isotopes, is believed to be small because the various data were taken from experiments which had a reactor residence time ranging from a few months to three years.

Of particular interest is the change in the Nd isotopic ratios and fission yields within a given reactor. This is shown for the measurements on EBR-II samples which were irradiated in row 4, row 8, and the axial blanket and amplifies the need for spectral information with respect to fission yields. Also to be noted is the significant change in the ,ield of 154Sm. This should be considered by those experimenters conducting isotope correlation studies with high mass number fission products or investigating the use of 155Eu as a burnup monitor for fast reactor fuels.

It is recognized that this study is still in the development stage and is based only on limited  235 U fission yield data, but it is believed to be the first time that such a correlation of relative yields with neutron energy has been generated and that the following observations can be drawn from the data.

- 1. For the most exacting burnup measurement, no one set of "fast reactor fission yields" exists which is applicable to all fast reactors.
- 2. Every effort should be made to define the neutron spectra associated with new experimental yield data.
- 3. Future fast fission yield compilations should not consist of average data from different experiments and more de-

tailed compilations as a function of neutron energy will be required.

The above listed comments represent the major plea that the users of fission yield data for the determination of burnup have to make to this conference, to the measurers of new fission yield data, and to the compilers of the data.

Additional observations pertaining specifically to the determination of burnup which can be made from the  $^{143}Nd/^{150}Nd$  ratio data for  $^{235}U$  (Fig. 2) are:

- 1. That conceivably, the measured isotopic ratio of unspiked Nd can give an indication of the neutron environment associated with the particular sample being analyzed.
- 2. That based on the measured isotopic ratios, it may then be possible to select the most appropriate fission yield value for the calculation of burnup.

It is realized that for fast reactor fuels having several sources of fission, these ratios may be more complex; however, studies with plutonium fast yield data show the existence of similar changes in the Nd ratios with neutron energy although they are not as great. Considering a fuel like that proposed for the USA fast breeder program which has a recycled-Pu to U ratio of  $\sim 1/3$ , it can be shown that from 75-85% of the fissions are due to  239 Pu (Table I). Hence, it may still be possible to use certain selected isotopic ratios to characterize the neutron environment.

The comparison study of the ²³⁵U relative isotopic fast fission yield data has been extended to all of the major elements in the mass yield curve. Continually changing ratios with neutron energy are observed for those elements on the wings and valley of the mass yield curve with little change for those elements on the peaks. These data [35] will be discussed in a separate paper at this conference.

## 3.2. Fission product nuclear decay constants

When using a radioactive isotope to measure burnup, it is very likely that the major error is associated with the nuclear decay constants ( $\beta^-,\gamma^-$  branching,  $\gamma$ -ray intensities, etc.), and may exceed that of the fission yield. Although considerable work has been done in this area, significant uncertainties are still associated with these decay constants.

An interesting comparison of burnup values derived from the  $\gamma$ -ray analysis of small irradiated samples of  235 U using several different nuclides has been made by Tasaka and Sasamoto [14]. In this study using published gamma ray intensities, differences in burnups of 10 relative percent were observed. Subsequently, they recommended a new set of gamma ray abundance values for several of the nuclides, but one can always question whether the observed errors were due entirely to this source.

It is preferred that gamma ray intensities or abundances be known to 1 relative percent for the major gamma rays of the most widely used fission product monitors. These are 95Zr-Nb, 106Ru, 137Cs, 140Ba-La, 144Ce, 144Pr, 154Eu, and 155Eu. One method of minimizing this error is to use "standard" sources of the gamma emitter being measured.



Fig. 3 Fission yield variations with neutron energy.

This technique is satisfactory if both sources are of the same mass and configuration; however, using a point source to calibrate a fuel rod scanner is unacceptable, unless a portion of the standard is introduced into the matrix being measured.

One highly recommended procedure for preparing standards of the longer lived (>30 days) radioisotopes is isotope dilution mass spectrometry [39] where the standard is calibrated directly in terms of atoms. Hence, all nuclear data errors except those associated with the fission yield and half-life are eliminated.

### 3.3. Fission product capture cross sections

In any method, destructive or non-destructive, for the determination of burnup based on the measurement of a fission product monitor, the number of atoms of the fission product monitor is used to measure the number of fissions. In the ideal case, the number of fissions is directly proportional to the number of fission product monitor atoms. However, in the real case, the number of fission product atoms may be altered by the fission products capturing neutrons. Both burnin and burnout of the fission product monitor must be considered.

#### 3.3.1. Thermal neutron spectra

For thermal neutron irradiations, the problem of fission product burnout can be minimized by selecting a monitor whose capture cross section is small (a few barns) compared to the relatively large (500 barns or greater) fission cross section of the fissioning isotope. The small neutron capture cross section of 148Nd was a primary consideration in selecting it as a reliable thermal burnup monitor; how-ever, the burnin reaction from 11.1d  147 Nd was not seriously considered because of the relatively short half-life and a poorly known capture cross section. The results of recent burnup analyses in our laboratory for fuel samples from a high flux reactor show an abnormally high ¹⁴⁸Nd/¹⁵⁰Nd ratio which increased with burnup. This ratio should have remained constant if there were no significant burnin or burnout reactions. Our conclusion is that the capture cross section for 11.1d ¹⁴⁷Nd may be in the region of 200-300 b and that for high flux irradiations (which are typical in fuel development studies) it is very probable that a burnup measurement based on 148Nd could be biased high. To estimate the magnitude of this bias, the effect of a high ¹⁴⁷Nd capture cross section upon a burnup measurement based on a ¹⁴⁸Nd analysis was calculated. For this calculation, fully enriched 235U fuel and a flux of  $5 \times 10^{14} n/cm^2 \cdot s$  were used. The results are given in Figure 4. A 10% high bias can be a serious problem in fuel development studies, especially if the data are being used to design future fuels with high power ratings. Thus, improved cross section data for ¹⁴⁷Nd are urgently needed.

The other major requests for improved fission product cross section data are primarily associated with the isotope correlation studies. Included are requests for capture cross section data for  83 Kr,  131 Xe,  133 Xe,  133 mXe,  133 Cs,  134 Cs,  141 Pr,  153 Sm,  153 Eu, and  154 Eu.

#### 3.3.2. Fast neutron spectra

For fast reactors, the problem of fission product neutron capture



Fig. 4 Effect of high ¹⁴⁷Nd cross sections on ¹⁴⁷Nd burnup values.

can potentially be more serious than for thermal reactors. The fission product cross sections may be as high as several hundred millibarns and the fission cross sections for the major fissioning isotopes are about 2000 millibarns; hence, the ratio of these cross sections can be greater than in thermal reactors. Fission product burnout as a function of burnup in fast reactor spectra is shown in Figure 5 for fission product capture cross sections ranging from 50 to 250 millibarns. This demonstrates the need to obtain accurate fission product capture cross sections in a fast neutron spectrum to meet the goal of 1-2% accurate burnup determinations.



Fig. 5 Fission product burnout.

# 3.4. Heavy element neutron capture cross sections

The primary needs for more accurate heavy element capture cross section data are: 1) for the isotopes  236 U,  238 U, and  242 Pu which when capturing a neutron, form a short lived isotope and decay to a different element and 2) for  240 Pu which can capture to form  241 Pu which subsequently decays with a half-life that may have an uncertainty of 5-10%. The magnitude of the error introduced into a resulting burnup value varies with the fuel composition, isotopic enrichment, neutron spectra, and irradiation history. For the most exacting work, a separate analysis of the sample is required to quantify the amount of decay product present. The ability to accurately calculate this value would result in considerable savings in costly analytical time.

## 3.5. Capture-to-fission ratio, $\alpha$

This value is primarily used in calculating burnup via the heavy element isotope ratio method (Section 2.1.2). Many accurate measurements of  $\alpha$  have been made and it is well known that  $\alpha$  can vary widely with neutron energy.

For some thermal reactors, core averaged values have been measured which can be used when large segments of the core are dissolved and analyzed. For small individual samples, a knowledge of the neutron distribution and temperature is required to arrive at an accurate value.

The capture-to-fission ratio for  233 U,  235 U,  238 U,  239 Pu, 240 Pu, and  242 Pu has been measured for several different locations in the fast reactor EBR-II [40]. Variations in  $\alpha$  of up to a factor of 10 have been observed within the core of EBR-II.

Thus, requesting more accurate  $\alpha$  data for use in the calculation of burnup does not appear practical because the values are too spectrum dependent, and generally, the user does not have sufficient information concerning the spectrum to make a reliable estimate of  $\alpha$ .

## 3.6. Half-lives

For the absolute determination of burnup using a radioactive nuclide, the uncertainty in the half-life will introduce varying degrees of uncertainty in the final result depending upon the magnitude of the in-pile and out-of-pile decay corrections. Eder and Lammer [9] have shown for existing half-life values that if the decay time does not exceed approximately one half-life of the nuclide being measured that the error from this source will rarely exceed 1%. In general, the half-life values for most of the major radioactive fission product burnup monitors are known with sufficient accuracy that new measurements are considered to be of low priority compared to decay scheme and fission yield requirements.

The error in the half-life is minimized when relative burnup measurements are conducted and eliminated when stable fission products are used as the monitors.

#### 4. SUMMARY

The fission product nuclear data requirements for the determination of burnup by both destructive and non-destructive methods have been reviewed. At this time, the greatest need for improved fundamental data is for fast reactor fission yields as a function of neutron energy, and it is strongly recommended that these data be associated with a well defined neutron spectrum. For thermal reactors, the current yield data appear adequate. When radioactive fission products are measured, the major error in the final burnup value arises from the uncertainty in the nuclear decay schemes. New data requirements with assigned priorities are listed in Table III.

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	NU	ICLEA	R DATA REQUIRE	MENT	STATUS FUTURE NEEDS	PRIORITY
I.	FIS	SION The 1.	YIELDS rmal Reactors General:	233U 235U 239Pu 241Pu	2-3% For long-lived and stable nuclides which comprise major fraction 1-2% of mass yield curve. 2-3% Adequate - not practical to initiate new extensive measurement programs.	Low
		2.	Specific: ¹⁰³ Ru, ¹⁰⁶ Ru, ¹⁴¹ Ce, ¹⁴⁷ Nd,	¹⁴⁰ Ba-La 153 _{Sm}	Variable from 2-5% These shorter-lived nuclides are used in radiochemical measurements for burnup and the yield of the stable end member of the chain is not easily measured. Desire 1-2% values. Must have a good technique to measure the number of fissions to establish reliable absolute yield data.	Medium
			¹³⁴ Cs, ¹⁴² Pr,	¹⁵⁴ Eu	Unknown or with very large errors. These are shielded nuclides with very low yields.	Medium

TABLE III. SUMMARY-NUCLEAR DATA REQUIREMENTS - STATUS - FUTURE NEEDS FOR FUEL BURNUP MEASUREMENTS

	NUCLEAR DATA REQUIREMENT	STATUS	FUTURE NEEDS	PRIORITY
I.	FISSION YIELDS (Cont'd)			
	B. Fast Reactors			
	1. General: ²³² Th	Poor	3-5%	Medium
	²³³ U	Not available	1-2% as function neutron spec	tra High
	235 _U	~3% for very hard spectrum	1-2% as function neutron spec	tra High
	238 _U	$\sim$ 15% undefined spectrum	3~5%	High
	239 _{Pu}	${\sim}3\%$ for very hard spectrum	1-2% as function neutron spec	tra High
	240 _{Pu}	Not available	3-5%	High
	241 _{Pu}	Not available	1-2% as function neutron spec	tra High
	²⁴² Pu	Not available	5-10%	Medium
	237 _{Np}	Not available	5-10%	Medium
	²⁴¹ Am	Not available	5-10%	Medium
	2. Specific: Nd isotopes The yields for the Nd iso topes are of particular importance because they will probably be the pre- ferred monitor and the isotopic composition may define the neutron spectr	Several measurements but most in poorly defined spectra. Yields and isotopic composi- tion change with energy.	1-2% yield values in well defined spectra. Isotopic composition to 0.5% for well defined spectra.	High

# TABLE III. SUMMARY-NUCLEAR DATA REQUIREMENTS - STATUS - FUTURE NEEDS FOR FUEL BURNUP MEASUREMENTS (Cont'd)

	NUCLEAR DATA REQUIREMENT	STATUS	FUTURE NEEDS	PRIORITY
ΤТ.	DECAY SCHEMES			
	A. Fission Products			
	¹³⁷ Cs, ¹⁴⁰ Ba-La, ¹⁴¹ Ce, ¹⁴⁴ Ce, ¹⁴⁴ Pr, ¹⁴⁷ Nd, ¹⁵⁴ Eu, ¹⁵⁵ Eu	Variable	l% uncertainty required for major γ-rays	High
	B. Heavy Elements	Adequate	Adequate	None
III.	NEUTRON CAPTURE CROSS SECTIONS			
	A. Fission Products			
	1. Thermal neutrons			
	¹⁴⁷ Nd of particular im- portance because may bias ¹⁴⁸ Nd results high.	Large uncertainty	10% or better	High
	⁸³ Kr, ¹³¹ Xe, ¹³³ Xe, ^{133m} Xe, 133 _{Cs, ¹³⁴Cs, ¹⁴¹Pr, ¹⁵³Sm, 153_{Eu,} ¹⁵⁴Eu.}	Variable - generally large	<3% for ¹³³ Cs, ¹³⁴ Cs, ¹⁴¹ Pr, ¹⁵³ Eu, ¹⁵⁴ Eu. ~10% for others adequate. All require resonance data	Medium
	2. Fast neutrons	Little information available.	(±10% for ≥ 50 b). ∿10% values as function of neutron energy for principal burnup monitors.	High

TABLE III. SUMMARY-NUCLEAR DATA REQUIREMENTS - STATUS - FUTURE NEEDS FOR FUEL BURNUP MEASUREMENTS (Cont'd)

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TABLE III. SUMMARY-NUCLEAR DATA REQUIREMENTS - STATUS - FU	UTURE NEEDS FOR FUEL BURNUP MEASUREMENTS (Co	mt'd)
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	NUCLEAR DATA REQUIREMENT	STATUS	FUTURE NEEDS	PRIORITY
III.	NEUTRON CAPTURE CROSS SECTIONS (Cont'd)			
	B. Heavy Nuclides			
	236 _U , 238 _U , ²⁴⁰ Pu, ²⁴² Pu	Differential data adequate although highly energy dependent.	Improved techniques for defining spectra such that proper value can be selected.	Low
IV.	CAPTURE-TO-FISSION RATIO			
	For all heavy nuclides	Differential data adequate although highly energy dependent.	Improved techniques for defining the neutron spectra such that repre- sentative values can be used.	Low
v.	HALF-LIVES	Generally adequate for major radioactive nuclides which are used as burnup monitors.	Improved data are always useful but not high priority item.	Low
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#### IMPORTANCE OF FISSION PRODUCT NUCLEAR DATA FOR SAFEGUARDS TECHNIQUES

#### C. Weitkamp

## Institut für Angewandte Kernphysik Kernforschungszentrum Karlsruhe, Federal Republic of Germany

#### ABSTRACT

The role of fission product nuclear data has been reviewed for the three important categories of safeguards techniques based upon the fission process: fresh fuel assay, nondestructive measurement of spent fuel by gamma spectrometry, and correlations of stable fission products with characteristic data of the fuel and its history.

Fresh fuel assay procedures are generally calibrated by standard samples; the needs of FPND for purposes such as instrument optimization, reduction of effort or improvement of accuracy for calibration procedures, or feasibility studies of new methods are neither very pressing nor, at the present time, quantitatively defined by sensitivity studies etc.

Spent fuel investigation by nondestructive gamma spectrometry has until now mainly been used for verification purposes; its quantitative use has been limited by FPND accuracies, mainly yields, half-lives, capture cross sections and resonance integrals, and gamma-ray intensities. Improvement of existing FPND would be helpful, but as spent fuel element accountancy is done digitally, the resulting improvement of accuracy would be more valuable to the reactor operator than to the safeguarding authorities.

Correlation techniques are an important means of verification of reprocessing input analyses. Whereas heavy isotope correlations have already been used and investigated extensively, correlations involving stable fission products and their ratios are only now becoming routine. The technique has so far exclusively been based upon empirical data with no utilization of FPND. The virtue of precise FPND is in the calculation of fission product correlations which are valuable for the prediction of the scope and limitations of various correlations. These calculations have often been unsuccessful, and better FPND are likely to improve the situation.

#### 1. INTRODUCTION

Soon after the potential hazards associated with the diversion of fissionable material from authorized uses in the growing field of nuclear energy had been recognized, a worldwide system for the detection and prevention of such diversion was developed which is now generally referred to as nuclear materials safeguards. Although safeguards procedures /1/ vary greatly according to the level of responsibility, material, and external conditions, material accountancy and the requirement for the closure of material balances is a common feature at all stages. Because the material undergoes constant change in the different facilities of the nuclear fuel cycle, frequent measurement of its flow and inventory is necessary. In addition to classical chemical and physical methods techniques for the determination of nuclear material have been developed that make use of its nuclear properties. It appears logical that among all physical phenomena utilized neutron-induced fission itself plays an important part. The connection of nuclear data with safeguards techniques has been one of the topi's of a recent international conference, and a number of contributions /2 - 5 have dealt with that somewhat more general subject. It is the purpose of this paper to review the particular role of fission product nuclear data (FPND) with respect to safeguards techniques, and thus contribute to a basis for recommendations as to the measurement, compilation and dissemination of FPND for use in practical applications.

#### 2. BORDERLINE BETWEEN FPND AND NON-FISSION-PRODUCT NUCLEAR DATA

It seems as if a clear-cut definition of which categories of nuclear data are FPND and which are not has not yet been given. Physically it appears logical to define as FPND all data needed or useful for a description of the processes that occur after the fissioning nucleus has lost its identity, and correlations of such data with the circumstances of its formation. Traditionally, however, fission products have been considered as such only if their half life exceeds some macroscopic value of the order of 1 second. The reason for this has been the limitation of early measurement, separation and identification techniques. For the purpose of this review I will generally adhere to the latter definition, but where necessary will mention needs for data pertaining to a shorter time range.

#### 3. SAFEGUARDS MEASUREMENT TECHNIQUES: GENERAL CONSIDERATIONS

Safeguards measurement techniques are conveniently divided in two categories according to whether they are mainly used for fresh or for spent reactor fuel. A third technique makes use of inherent relationships between the quantities and ratios of certain isotopes; this so-called correlation technique is usually applied to the results of measurements on the dissolved fuel and will be treated separately.

The purpose of safeguards procedures developed for use on fresh fuel including scrap and waste is always the quantitative determination of an element, an isotope or a ratio of isotopes, sometimes the establishment of an upper limit for such a quantity. Although the devices to be used are subject to the usual limitations as to measuring time etc. and are sometimes expected to provide additional information for other purposes such as quality assurance, they must fulfill high standards as to precision, reliability and overall accuracy. The figure aimed at is usually of the order of 0.3 % total error. As FPND are seldom known to that accuracy, this means that methods for fresh fuel must not directly depend upon FPND, and they indeed don't. Instead, instruments and methods are generally calibrated with respect to standard samples the production, analysis and proper use of which is not a trivial problem.

Methods for spent fuel, on the other hand, in addition to the measurement of quantities or ratios of fissionable material, must also serve purposes connected with the history of the fuel such as the determination of flux level, neutron spectrum, burnup, plutonium-to-uranium fission ratio, cooling time etc. This information will often already be available from the reactor operator, and the aim of the development of a method is usually not the ultimate in precision, but reasonable independence from unknown or uninteresting physical quantities in order to render the results dependable, generally applicable and thereby acceptable to operators and inspectors. Precise determination of fuel quantities is of secondary importance because bookkeeping in reactors including spent fuel storage pools is usually done by item, i.e. fuel pin, fuel element etc.

It is only after dissolution in the input accountability tank of the reprocessing plant that the change is made from so-called digital accountancy to open accountancy. Here correlation techniques play an important role both for the safeguarding agency who is interested in a means of verification of the wet chemical and other direct determination of fissionable material, and the owner of the fuel who needs additional information about data important for the operation of the reactor such as the burnup level reached.

# 4. FPND DEPENDENCE OF VARIOUS METHODS FOR FRESH FUEL ASSAY

Techniques for the determination of fresh fuel only depend upon FPND if the physical phenomenon utilized for the assay is the fission process. This excludes many methods and instruments from further consideration. Also I will not discuss methods that have been given little consideration until now or that are unlikely to be used in the near future because of cost, complexity, measurement time or simply better alternatives.Examples may be the use of neutron spectrometry for delayed neutron emission  $\frac{6}{7}$ or resonance neutron transmission measurements  $\frac{78}{78}$ .

As mentioned above, fresh fuel methods do not directly depend upon FPND, but the precise knowledge of such data is sometimes helpful or interesting for a variety of reasons.One of them may be the FPND dependence of correction terms that are difficult to measure. Another can be a reduction of the calibration effort, both in terms of a reduced number of standards and a simpler or more accurate calibration procedure; and sometimes optimization of the design of an instrument of proven operability requires the knowledge of FPND.

In order to explain what this means I will briefly describe three methods, one for each of the categories above. I will then just mention a few more methods where FPND would be interesting to have, but where the immediate benefit is at least questionable. After that, I will show an example where FPND are needed for the data reduction, i.e. computation of correction terms, but present accuracies are completely adequate; and finally, for the sake of completeness, I will mention a few more methods which, although based on the fission process, require no FPND at all.

# 4.1 252 Cf Assay System for FBR Fuel Pins

One of the finest instruments presently in use for the assay of fuel pins for fast breeder reactors is the  252 Cf assay system developed by the Los Alamos Scientific Laboratory / 9 / . Of the different individual measuring devices included in the instrument I will only discuss the one for the determination of the total content of fissile material in a pin which is the most important one from a safeguards point of view. The technique is based upon the activation of the rod by carefully spectrum-tailored  252 Cf neutrons and subsequent measurement of the gross  $\gamma$  rays from fission products in the energy range between 1.2 and 2.7 MeV. The system is being calibrated about once a day by means of standard rods of known quantity and composition.

Although the instrument is equipped with an elaborate data reduction system that automatically corrects for background from passive gammas, source decay etc. both in the calibration and assay runs, there seems to be a systematic long-term change in response superimposed on the final results that could be due to incomplete compensation for activation buildup in the standard rods. A complete library of fission product yields, half lives and  $\gamma$ -ray energies and intensities could probably be used for a quick check of the magnitude of the effect to be expected, and possibly help to find some means for its compensation.

## 4.2 Assay of LWR Pins by Activation in a Reactor and $\gamma$ Spectrometry

A method for simultaneous assay of  235 U and fissionable plutonium in LWR pins consists in the activation of the pins in a reactor followed by  $\gamma$ -ray spectrometry / 10 /. The method has so far not been used routinely although it is relatively fast, simple and inexpensive if a small reactor is available. If the irradiation, waiting, and counting times which are all of the order of a few minutes are precisely reproduced, spectroscopy of the delayed  $\gamma$  rays with a Ge(Li) detector offers enough discrimination power to determine the amount of the individual fissile isotopes. Again calibration is done with standard samples; the amount  $Q_i$  of isotope i is computed from the peak areas  $P_p$  via the relation

$$Q_{i} = \sum_{k=1}^{n} A_{ik} P_{k} \qquad i = 1...m, m \leq n$$

where the constants  $A_{ik}$  can be, and have until now been determined from a measurement of the standards. This calibration procedure involves the inversion of matrices all elements of which are subject to statistical errors, and can therefore become quite inaccurate, particularly if more than two isotopes are to be determined. The precise knowledge of the yields, half lives and  $\gamma$  intensities of the fission products involved (together with the fission cross sections of the heavy isotopes) determines the ratios  $A_{ik}/A_{ik}$ , reducing the above equation to

$$Q_{i} = \sum_{k=1}^{n} \lambda_{ik} \lambda_{k} P_{k}$$

where the  $\lambda_{ik}$  are all known and only n calibration parameters A have to be determined (one for each peak), instead of n × m (i.e. one for each peak and each isotope). Until now, however, although there exist excellent measurements of delayed Y rays in the time range down to about 5 minutes, even the most prominent of the  $\gamma$ -ray peaks of these short-lived fission products could not be assigned to individual nuclides / 11, 12/.

Activation buildup in the standards, as mentioned in the discussion of the previous method, is also present. As long as this effect only requires a different determination of the background under the peaks, the resulting error should be negligible. Care must be taken, however, that there occur no errors due to count-rate effects, and that there is no buildup of long-lived peaks close to the peaks of interest that introduce systematic errors. Again, a comprehensive library of precise FPND, particularly for the longer-lived fission products ( $T_{1/2} > 1$  day), could be helpful for a computational a-priori investigation of the problem.

## 4.3 Isotopic Source Assay System with Coincident Particle Detection

An active system for the assay of fissionable material in a wide variety of chemical compositions and geometric shapes that has been commercially available for some years and had originally been developed by Gulf Radiation Technology / 13,14 / uses a  252 Cf neutron source similar to the system described in paragraph 4.1. The secondary particles detected, however, are neutrons and quasi-prompt  $\gamma$  rays. In order to get the best signal-tobackground-ratio, a selectable number of detectors is required to fire simultaneously for an event to be registered. Typical settings of the coincidence requirements are, e.g., two-out-of-four and three-out-of-four, for a frequently encountered version. It is obvious that the design of the instrument, i.e. source strength, number, size, efficiency and shielding of detectors etc. depends upon the number, nature and energy of the particles emitted "simultaneously" in a fission event. Simultaneously, in this context, means that the particles can be registered using standard detectors and coincidence circuits the resolving time of which is of the order of 20 nanoseconds. It does not mean that the particles have to be generated simultaneously on a nuclear time scale which is much finer, or even by advanced laboratory equipment which is more than 3 orders of magnitude more sensitive, but of course not suited for routine work.

For neutrons, the distribution of multiplicities is moderately well known, quite good for thermal-neutron induced fission, poorly for higher neutron energies. For  $\gamma$  rays, practically no measurements have been made. In addition, whereas both in the determination and utilization of neutron multiplicities initial neutron energy is unimportant as long as the neutrons are thermalized prior to detection, there is almost always a strong energydependence in the detection of the  $\gamma$  rays, and certainly an energy correlation of their multiplicity. Although data of this kind, as outlined in chapter 2, may not directly belong to the subject of the panel, they appear to be of sufficient importance to be mentioned, and the needs for their measurement should not go unnoticed.

#### 4.4 Miscallaneous Methods for which FPND are of Interest

There are a few other methods for which a better or more complete knowledge of FPND could simplify operations or provide possibilities for additional checkout or improvement of the apparatus. Such data are the multiplicity distribution of prompt neutrons emitted in fission of ²³⁵U, ²³⁹Pu and ²⁴¹Pu by subthreshold (e.g. Pu-Li) neutrons / 15_/, or in spontaneous fission of ²³⁸Pu, ²⁴⁹Pu and ²⁴²Pu. A detailed discussion of this question has been given in / 5_/ and need not be repeated here. Data interesting for the development and application of methods based on photofission are the variation of  $\bar{\nu}$ , the average number of neutrons, for subthreshold energies / 16_/. Both types of data, however, may or may not be considered as FPND (cf. chapter 2). Another category of data whose identity as FPND is questionable are the half lives, yields,  $\gamma$ -ray energies and  $\gamma$ -ray intensities of fission products in the time range below one second down to perhaps 1 µs. It seems as if some measurements had been done in this field, but prospective users of the data complain about the lack of a coherent and comprehensive review of the experimental material / 16 /.

4.5 Trace Analysis of ²³⁵U and ²³⁹Pu by Reactor Activation and γ-Ray Spectrometry of Noble Gas Decay Products

The analysis of microgram amounts of fissionable isotopes as developed at the Kernforschungszentrum Karlsruhe / 17,18 / is discussed in this chapter because it fulfills the general criteria outlined in chapter 3,not because of its limitation to fresh fuel; in fact, it has successfully been used on hot dissolver solutions from reprocessing plants.

The method is based on thermal-neutron induced fission of  235 U and  239 Pu in a reactor. The noble fission gases are adsorbed on a charcoal filter shortly after the irradiation; the residual gas is removed from the filter after about 3.5 hours. The remaining non-volatile decay products of  88 Kr and  138 Xe,  88 Rb and  138 Cs, are then measured with a Ge(Li) detector utilizing their intense  $\gamma$  rays at 1834 and 1438 keV, respectively.

The area P of the photopeaks of these lines is proportional to the (unknown) number N of heavy atoms and related to the flux  $\Phi$ , fission cross section  $\sigma$ , yield Y and decay constants  $\lambda_1$  and  $\lambda_2$  of the parent (noble gas) and daughter (alkali) isotopes via the relation

P =	N	ଡ଼ୢୢଡ଼	Y	(1-e ^{$-\lambda_1 t_1$} )e ^{$\lambda_1 t_2$}	$\frac{\lambda_2}{\lambda_2 - \lambda_1}$	$(e^{-\lambda_1 t_3}-$	$e^{-\lambda_2 t_3} e^{-\lambda_2 t_4}$	εI	$\frac{1-e^{-\lambda_2 t_5}}{\lambda_2 t_5}$	t ₆
-----	---	--------	---	--------------------------------------------------------------------------------------	-------------------------------------------	------------------------	-----------------------------------------	----	----------------------------------------------	----------------

where t₁ is the irradiation time, t₂ the waiting (transfer) time, t₃ the adsorption time, t₄ the time between desorption and the beginning of the measurement, t₅ and t₆ the true and live measurement times, and  $\varepsilon$  and I the peak efficiency of the detector and intensity of the  $\gamma$  ray. There are of course two such equations of identical structure, but with different constants  $\sigma$ , Y,  $\varepsilon$ , I,  $\lambda_1$ ,  $\lambda_2$  for ²³⁵U and ²³⁹Pu.

Again, calibration is done with accurately prepared standard samples, containing a small quantity of cobalt as a flux monitor, and does not depend upon FPND as long as irradiation, adsorption, desorption, and measurement conditions (esp. times) remain unchanged. This, however, is often not the case, and the procedure is to calibrate the products  $\sigma Y \varepsilon I$  for a given set of time parameters and flux and use the above equation with the measured values of  $t_1$  through  $t_6$  and  $\Phi$ .

Although the correction terms for values of the time parameters that differ from the ones used in the calibration depend directly upon FPND, this effect is now shown to be negligible. The results of an FPND sensitivity study made for this purpose are summarized in Table I. Uncertainties of the half lives of the four isotopes involved have been assumed to be 1 % which is slightly larger than the currently adopted error for ⁸⁸Kr and ⁸⁸Rb / 19 / and may also be realistic for the two mass-138 nuclides. As can be seen from table I, half life errors will hardly contribute more than 0,25 % if the prescribed procedure, particularly as to the duration of the adsorption, is closely followed. The overall accuracy of the method for separate assay of each of the two fissionable isotopes in mixtures (mixing ratios between 1 : 3 and 3 : 1 and total quantitles between 6 and 20 µg) is now 3 % only, therefore the error from FPND uncertainties is more than an order of magnitude smaller than all other errors, and there is no need for an improvement of our knowledge of the half-life values of the four fission products utilized.

# 4.6 Other FPND-Independent Methods for Fresh Fuel Assay

Perhaps the most important active method for the assay of fissionable material in fresh fuel is the measurement of delayed neutrons following irradiation of the sample with a short burst of neutrons / 20,21,22 /. Although knowledge of the time distribution of the delayed neutrons and the prompt-to-delayed-neutron ratio are very valuable, the precision to which any kind of FPND are known does not presert a limitation of the method. The same statement can be made for the reactivity method where the reactivity change of a subcritical assembly upon insertion of the sample is measured / 23 /, for the interrogation of fuel pins or fuel elements with subthreshold neutrons from a lead pile / 24,25 / or Sb-Be source / 26,27 / and detection of fast fission neutrons, and for all other major methods currently applied or under development that are based on neutron-induced,  $\gamma$ -induced, or spontaneous fission of heavy isotopes.

#### 5. FPND NEEDS FOR NONDESTRUCTIVE INVESTIGATION OF SPENT FUEL

For spent fuel discharged from a reactor the accessibility for measurement is even worse than for fresh fuel in fabricated fuel elements, and if any knowledge about properties of the fuel is required prior to reprocessing of the element, it can practically only be obtained by nondestructive methods. Although a few other methods have been developed for the investigation of hot reactor fuel - active neutron interrogation with delayed neutron counting / 28 /, gamma and resonance neutron transmission measurements / 29, 7 / and calorimetry / 30, 31 / -, the only important tech-

						Γ
J	assumed value for calibration	actual range of values used	maximum percent difference	error componen assumption that the used fo	it caused by $\Delta t_i$ under the ire is a 1 % error in the ir the half life of	alue
	minutes	minutes	∆t. 1	⁸⁸ Kr (2.80h) ⁸⁸ Rb (17.	8m) $\left  \begin{array}{c} 138 \\ 138 \\ \text{Xe} (14.2m) \end{array} \right ^{138} \text{Cs} (14.2m) $	2.3m)
	10	5 - 10	-50%/ 0	- 0.01 %	- 0.10 %	
	4	4 - 5	0 /+25%	+ 0.004%	+ 0.05 %	
	214	214 - 224	0 /+ 5%	+ 0.04 % + 0.00	28   + 0	22 %
بر ۲	4	4 - 11	-438/+578	-,12%/+.	15%	\$60.+
•	11	4.17-20.58	-62%/+82%		15%	\$60.+
cas ight	e" combinat: er assumed ( literature (	ion of paramet to be both 1% values	cers; half larger	25%/+.35%	25%/+.45%	

TABLE I. RESULTS OF A STUDY OF THE SENSITIVITY OF A METHOD FOR TRACE ANALYSIS OF ²³⁵U AND ²³⁹Pu BY REACTOR ACTIVATION AND GAMMA-RAY SPECTROMETRY WITH RESPECT TO ERRORS OF THE HALF LIVES OF ⁸⁸Kr, ⁸⁸Rb, ¹³⁸xe AND ¹³⁸Cs

nique in wide use now is spectroscopy of the intense  $\gamma$  rays from fission products, almost exclusively with semiconductor  $\gamma$ -ray spectrometers (Ge(Li) or intrinsic germanium), although NaI(T1) detectors have also been used / 32,33/.

Whereas the calibration of methods and instruments for fresh fuel assay is basically done by comparison with accurately prepared or well-analyzed standard samples which do not change in time except for minor changes in isotopic composition of plutonium, grow-in of americium, and occasional weak activation by the calibration process itself, the preparation of such standards for spent fuel is obviously not possible. Unless there is a straightforward check of the validity of a method as, e.g., in the case of cooling time determination, the best thing one can expect to get is a detailed report of the results of the analysis after dissolution. Even then the data are not always comparable because batches in the dissolution are usually quite large, whereas single pins of portions of pins can be investigated by  $\gamma$  spectrometry, and also because the material may undergo considerable change in composition between the two determinations.

This means that  $\gamma$  spectrometric measurements of spent fuel rely heavily on nuclear (and some non-nunclear) data of fission products, and the question is not only if and which FPND are relevant to the different methods, but also whether existing FPMD are sufficiently accurate. The answer to this question is seldom possible without extensive investigation. The reason for this is that the interpretation of the measured values for certain peak areas or peak area ratios often requires the use of large computer codes that provide for effects of parameters such as neutron spectrum, coolant void factor, temperature, history of control rod operation etc. Indeed, few investigations of the effect of FPND uncertainties upon the results have so far been carried out. Although the announcement of the present meeting has stimulated very promising work in the field, a final answer as to the detailed FPND requirements can only be given at a later date. Whereas data requirements for the assay of fresh fuel - however few there are - are at least quantitatively different from those for other purposes such as waste management or reactor design and operation, the requirements for spent fuel rethods including correlation techniques (cf. chapter 6) will be very similar to the requirements of other applications in which essentially the same type of codes can be, or are being used. In fact, there is more or less overlap with the subjects of review papers 2, 3, 4, 5 and 7 of this panel; in particular, paper 5 is devoted to burnup determinations which is one of the major objectives of nondestructive post-irradiation analysis and one of the important quantities in correlation techniques as well. Therefore the burnup question will not be treated here, or just briefly be mentioned for the sake of completeness.

In the discussion of the determination of the important quantities the different steps will be described in the same order in which they would have to be taken in an actual determination, largely following the procedure outlined in Ref. /3/. In order to produce consistent results an iterative procedure is usually applied.

# 5.1 Cooling Time

Cooling times, if not available as part of the operation history of the reactor, can be determined by  $\gamma$  spectrometry from the ratio of peak areas of fision products with different half lives. This procedure, however, is not without problems. First, the reactor must have been operated at constant power for a time long enough that both fission products are in equilibrium before shutdown. This limits the method to the use of relatively short-lived isotopes such as  $1^{49}$ Ba (12.8 d) -  $1^{40}$ La (40.2 h) / 3, 33, 34, 35 /,  $1^{31}$ I (8.05 d) / 3 /,  $1^{41}$ Ce (32.5 d) / 3, 34/, and  $9^{5}$ Zr (64.0 d) -  $9^{5}$ Nb(35.0 d) / 34,35 /; but  $1^{5}$  Ce (284 d) -  $1^{46}$ Pr (17.3 m) / 35 / and even  $1^{37}$ Cs (30.0 a) / 35 / have also been utilized. Second, the use of genetically related isotopes

which is conceptually the best method as long as the independent yield of the daughter is negligible relative to the cumulative yield of the parent is often not very sensitive. As the ratio  $R_0$  of the parent-to-daughter activity at time 0 (i.e. immediately after shutdown from constant power) and at infinite time,  $R_m$ , are related by the equation

$$R_{\infty} = R_0 \quad \{1 - / T_{1/2} (daughter) / T_{1/2} (parent) / \}$$

Ro and R_∞ differ only by a factor of 2.22 in the case of  95 Zr -  95 Nb, and only by 1.15 for  140 Ba -  140 La which is hardly adequate. Third, care must be taken that self-absorption factors are correctly taken into account. That practically eliminates the use of  141 Ce with a single  $\gamma$  ray at 145 keV. And fourth, perhaps worst of all, forward calculations of activity ratios vs cooling time have shown that the curves scatter by approximately + 15 % according to fuel composition and irradiation conditions, even if grow-in by capture is neglected and complete saturation at shutdown assumed / 34 /.

As half lives of the relevant isotopes are known to an accuracy of 0.05 to 0.2 percent and  $\gamma$ -ray intensities (or branching ratios) to about 1%, this means that FPND uncertainties are negligible, and existing data are fully adequate for age monitoring of spent fuel.

#### 5.2 Burnup

The cooling time of the fuel being known, burnup can be determined from an absolute measurement of the amount of certain radioactive isotopes. In order to be suited for burnup determinations except for very short irradiations times  $\int 36$ , an isotope has to be sufficiently long-lived, emit intense penetrating (>500 keV)  $\gamma$  rays, have a yield as independent as possible of fuel, spectrum, and other irradiation conditions, should not migrate under extreme temperature conditions, and must not be produced by processes other than fission nor burn out at high fuel burnup levels. Table II shows that three fission products or pairs of fission products are suited for the purpose; a more detailed discussion of their virtues and shortcomings is e.g. given in  $\int 38 / \text{ and } / 39 / \text{.}$  Where the requirement for an absolute determination could be met, excellent results have been obtained using 137Cs / 40, 41 /, but 144Ce -144Pr also seems to give satisfactory results / 35, 41 /.

Where absolute measurements by  $\gamma$ -ray spectrometry have not been possible ratios of isotopes that are produced proportional to different powers of the fluence have been used or proposed for investigation as burnup indicators. Among these are ratios of ¹³⁴Cs (2.05 a) and ¹⁵⁴Eu (16 a) as second-power and ¹³⁷Cs (30.0 a) and ¹⁴⁴Ce - ¹⁴⁴Pr (284 d) as first-power-isotopes / 41,42,43 /. However, these ratios vary considerably (for a given burnup level) with irradiation history, position of control rods, and cooling void fraction (in BWRs), and the relation

(secondary FP)(primary FP) = constant x burnup^k where k is of the order of 1.1 has been found to change so drastically in the outer parts of a BWR core that k had to be adjusted to a value of 0.5 in order to fit the measured data. This is attributed to spatial changes in the spectrum, but has not been understood quantitatively / 41 / .

Because burnup determination is the subject of an entire paper at the present panel, it does not seem appropriate to go into any further detail here. Nor will I touch upon the question of FPND requirements for burnup determination, although it seems obvious that a removal of some of the discrepancies between various yield determinations /44 / is highly desirable.

#### 5.3 Fluence

Unless special fluence-monitoring materials - isotopes not produced in fission, with reasonably large cross sections and long-lived daughters

Ϋ́Ρ	BLE II. FIS	SION PRODUCTS	USED FOR GAMMA-SPEC	TROMETRIC NOND	ESTRUCTIVE BURNUN	DETERMINATIONS
fission products	half live	as migration	percent yield [†] for thermal fission of ²³³ U ²³⁵ U ²³⁹ Pu	production by parent yield isotope ²	neutron capture: § half o life barns	burnout by neutron captures cross section o and resonance integral RI [¶]
⁹⁵ Zr- ⁹⁵ Nb 144Ce- ¹⁴⁴ Pr 137Cs	64.0d - 35 284 d - 17 30.0a	. Od no . 3m no Yes	6.27 6.45 4.90 4.52 5.42 3.85 6.12 6.27 6.48	⁹⁴ Zr 6.45 1 ⁴³ Pr 5.92 1 ³⁶ Ke 6.43	∞ 0.075 13.6d 89 ∞ 0.281	$\sigma = 1 b ({}^{9} {}^{5} Z x)$ , 4 b ( ${}^{9} {}^{6} N b$ ) $\sigma = 1 b ({}^{144} C e)$ , $RI = 2.2 b ({}^{144} C e)$ $\sigma = 0.110 b$
t evaluated f for therma weighted a f evaluated	values from L fission o werage of th values from	$f^{-37}_{-2350}$ , evalue he original di $f^{-44}_{-7}$ .	ated values from / 3 ata listed in / 37_/	7_7; the ¹³⁶ Xe 7 has been adop	chain yield bei' ted;	g obviously incorrect, the
TABLE III.	SENSITIVITY	OF THE COMPU	TED PRODUCTION OF ¹³ LISTED BEI	"Cs AND ¹³⁷ Cs OW, FROM / 46,	(IN PERCENT) TO 1 477+	N ERROR OF 1 % IN THE FPND
isotope F	ιτοάυςεα	sensitiv	lty to yields	sens	itivity to cross	sections
	ų,	1a1 _I 1 ³² T 0.0002 0.00	e 1 ^{3 3 m} xe ^{13 3} xe 13 0.0239 0.971	1 ³² X 0.00	e ¹³³ Xe ¹³³ C [.] 15 0.0124 1.00	¹³⁴ Cs 0 0.318
137 _C	ω	¹³⁴ Xe ¹³⁵ X	e ¹³⁵ Xe ¹³⁷ Cs 49 0,0070 0.984	¹³⁴ Xe ¹³⁴ C 0.0001 -	s ¹³⁵ Xe ¹³⁵ C	¹³⁶ Xe ¹³⁷ Cs 0.0103 ≈0.C1
t under the stainless burnup lev	following c steel cladd rel 40 000 M	onditions: PW ing 0.38 mm tl Wd/t U, cross	R, UO2 fuel, 3.1% er hick, moderator-to-f sections averaged c	rriched, pellet fuel volume rat vver spectrum a	diameter 8.9 mm io 1.6, moderato: t 12 000 MWd/t.	density 10.3 g/cm ³ , t temperature 285°C,

such as ¹⁰⁹Ag, ⁶⁴Zn and ⁵⁹Co - have been added to the fuel /45, the determination of the fluence, or time-integrated neutron flux, is only possible by measurement of the ratio of two isotopes the production of which is proportional to different powers of the flux  $\emptyset$ . This means practically that one of them has to be a fission product, the other one an isotope that is produced by neutron capture in a fission product, but must not be a fission product itself, and that both fission products have to have the same properties as those required for burnup determination (cf. paragraph 5.2, Table II). Long lifetime of the intermediate (capturing) fission product is a particularly important requirement because only then is the production of the daughter proportional to (f  $\emptyset$  dt)² where the integral is over the whole irradiation time.

It turns out that the only practical pair of nuclides is  $^{134}Cs - ^{137}Cs$ ; however, because the yields of both  $^{133}Cs$  and  $^{137}Cs$  vary with the fissioning isotope and the neutron spectrum, assumptions have to be made as to the fuel composition (such as pure  $^{235}U$  fissions) and spectrum (e.g., given ratio of epithermal to thermal flux). Then the fluence can be deduced from the measured ratios.

The sensitivity of the method to errors in cross sections and yields of the fission products involved has been studied in a recent calculation /46, 47 /; table III lists the sensitivities of the production rates of T³⁴Cs and ¹³⁷Cs with respect to the most important FPND. As expected, the yield, with a 1:1 correspondence to the production, is the only parameter of importance for ¹³⁷Cs; neutron capture in ¹³⁷Cs has no significant effect at a burnup level of 40 000 MWd/t. - For ¹³⁴Cs, both yield and cross section of ¹³³Cs enter directly into the result, but burnout is important enough to change the result by 1 % for 3 % error of the ¹³⁴Cs cross section.

This means that yields of ¹³³Cs and ¹³⁷Cs and capture cross sections of ¹³³Cs and ¹³⁴Cs are the important FPND. Whereas errors of current yield values vary between 0.5 % and 5 % for thermal fission of ²³³U, ²³⁵U, ²³⁹Pu and ²⁴¹Pu / 37 /, errors for fast yields are certainly at the upper end of this range, or somewhat larger. More important, however, are the uncertainties of the capture cross sections of ¹³³Cs and ¹³⁴Cs which are still in the 5 - 10 % region, and of which little is known about the variation with neutron energy / 3 /; it is the accuracy of these data that presently limits the accuracy of methods involving the use of calculated ¹³⁴Cs / ¹³⁷Cs ratios.

## 5.4 Irradiation Time

If the fluence is known, the irradiation time and average flux can be determined separately from the ratio of two isotopes one of which is not saturated under normal conditions (such as  $^{137}Cs$ ), and one of which clearly shows saturation effects (such as  $^{95}Zr - ^{95}Nb$ ). Nomographs for the determination of the irradiation time from known values of the fluence and  $^{95}Zr/^{137}Cs$  ratio have been computed /3/; although FPND sensitivity of the procedure has not been investigated it is clear from the absolute values of the FPND that only the yields of mass chains 95 and 137 and the half lives can be important. Half lives of  $^{95}Zr$ ,  $^{95}Nb$  and  $^{137}Cs$  are accurate to 0.2 %, 0.3 % and 0.7 % /44/ and thus perfectly adequate. As the sum of the mass numbers (95 + 137 + 2.5 = 234.5) is very close to the compound nucleus mass number for fission of both  $^{233}U$  and  $^{235}U$  the yields, although inaccurate to about 3 %, tend to change simultaneously with changes in the yield distribution and thus should have a small effect on the ratio of the two fission products. In addition, the effect of nonconstant power level of the reactor upon the  $^{95}Zr/^{137}Cs$  ratio is so drastic that no improvement of existing FPND seems to be necessary.

#### 5.5 Plutonium-to-Uranium Fission Ratio

An estimate of the ratio of fissions in plutonium to fissions in uranium can be obtained from the activity ratio of two fission products with markedly different yields for the two fissionable elements, with long half life and all the other properties already mentioned in paragraph 5.2.

The best pair of fission products for that purpose is  144 Ce (284.5 d) -  144 Pr (17.3m)/ 106 Ru (368.3 d) -  106 Rh(30 s), with yield ratios ( 144 Ce/ 106 Ru) of 17, 14, and 0.9 for  233 U,  235 U and  239 Pu thermal fission, respectively / 37/. The ratio R of  144 Pr/ 106 Rh activity decreases with an effective half life of 3.43 a; the error from the half-lives of  144 Ce(0.14 %) and  106 Ru(0.54%) is proportional to the cooling time and contributes to the computed value of R₀ (at shutdown) with only 0.50 %/year.

The ¹⁴⁴Pr/¹⁰⁶Rh ratio can best be used for irradiation times short with respect to the half lives, i.e. under one year, and appears to be reasonably sensitive to plutonium/uranium fission ratios between 0.05 and 1 and rather independent of the spectrum and of initial enrichment. For longer times, the situation remains relatively simple for pure uranium fuel as long as constant flux and quadratic increase of plutonium fissions can be assumed. Even then the computation of the plutonium-to-uranium fission ratio from the ¹⁴⁴Ce/¹⁰⁶Ru ratio at shutdown involves terms increasingly important with larger fluence that depend upon differences of yields. Although no quantitative investigation of the sensitivity with respect to FPND has been made, it is clear qualitatively that again yields are the limiting factor whereas accuracies of other FPND such as half lives and fission product cross sections are adequate.

Until now ¹⁰⁶Ru - ¹⁰⁶Rh has turned out to be the best plutonium fission monitor so far used / 3, 34, 35, 48 /, and further investigation of its potential has been suggested / 42 /. Attempts were also made to use other ratios such as ¹⁴⁰Ba(12.8d) - ^{T40}La(40.2h) / ¹¹¹Ag(7.5d) and ¹⁴⁰Ba - ¹⁴⁰La / ¹⁵⁶Eu(15.1d) / 34 /, but failed, probably because of too low absolute yields of ¹⁵⁶Eu and ¹¹¹Ag. The investigation of ¹⁵⁴Eu(8.5a) / ¹³⁷Cs(30.0a) has been proposed / 35 / because of the attractively long half lives involved. As ¹⁵⁴Eu is shielded by stable ¹⁵⁴Sm, it is primarily produced by capture in ¹⁵³Sm, with contributions from multiple capture in mass-152 down to mass-147 nuclides becoming increasingly important at high burnup levels / 3 /. The ratio of ¹⁵³Sm yields from thermal fission of ²³⁵U and ²³⁹Pu is 0.163% / 0.44%, or 1: 2.7; the production of ¹⁵³Eu from low-enriched uranium will therefore be proportional to almost the third power of the fluence even for the single-capture path, and to higher powers for the other modes of formation, and thus critically depend upon the spatial flux distribution and the fission product yields involved.

#### 5.6 Initial Enrichment

For low-enriched uranium fuel the initial enrichment can be computed from the ratio of plutonium-to-uranium fissions if the fluence, irradiation time and cross sections of the heavy isotopes involved are known. The calculations are easy if flux and initial enrichment are approximated by space-averaged values, and no time dependence of the flux is considered. For more refined calculations, the well-known large codes have to be used.

As the plutonium-to-uranium fission ratio is the quantity from which to start, the most important FPND are again the yields of mass chains 106 and 144; errors of half-lives and cross sections of fission products contribute little to the enrichment determination.

## 5.7 Remaining Fuel

The amount of remaining ²³⁵U can be calculated for reactors burning low-enriched uranium fuel if the burnup, plutonium-to-uranium fission ratio, and initial fuel are known.

For the determination of burnt-in plutonium an interesting method has also been proposed that consists essentially in a short (several hours') reirradiation of the cooled, spent fuel element followed by  $\gamma$  spectrometry. Although the effort of the reirradiation is considerable and not likely to be applicable to spent fuel investigations for safeguards purposes, there may be certain cases where the need for more accurate or more detailed information than obtainable by simple  $\gamma$  spectrometric means justifies such a procedure, and its potential ought to be more thoroughly investigated.

The method has been applied to fuel of the Bucharest VVR-S reactor at 10 % burnup (FIFA) after 600 days of cooling time / 49 /. The plutonium determination is quoted to be in error by 7 %. An analysis of the FPND dependence has also been made, but only with regard to the half life of ¹⁴⁰Ba, the short-lived fission product used in that particular case for the plutonium analysis. It was found that a half-life error of 1.5 % contributed 1.8 % uncertainty to the final result.

#### 5.8 General FPND Involved in Nondestructive Investigation of Spent Fuel

#### 5.8.1 Decay scheme data

Energies and intensities of gamma rays from longer-lived fission products are usually known well enough not to be a serious limitation of any of the methods listed above. Whereas a number of recent comprehensive compilations and evaluations exist of such data as yields and cross sections, the important decay scheme data (including, e.g. branching ratios of beta transitions to isomeric states) are still widely scattered in the literature. Therefore a number of laboratories produced their own private libraries, often suitably condensed, of the data required / 3, 47, 50 /. It is likely that publication of these libraries would be of general interest.

# 5.8.2 Gamma-Ray Attenuation Coefficients

Although  $\gamma$  ray self absorption and detector efficiency are important parameters, they are usually not determined from FPND. Detectors are calibrated using specially prepared  $\gamma$ -ray standards whose energy and source strength are very well known (better than 1 %) and for which self-absorption is negligible. Once the detector efficiency curve is known, self absorption correction factors for the fuel are best determined from known ratios of  $\gamma$ -ray intensities from the same isotopes. This procedure automatically corrects for  $\gamma$ -ray collimator effects etc. and only relies on relative intensities.

## 5.8.3 Migration

Fission products such as cesium, barium and ruthenium are known to migrate in the fuel at high temperatures and large temperature gradients /38,39 /. Whereas radial migration in fuel rods can be overcome by suitable design of the  $\gamma$ -spectrometric equipment, axial migration is a more severe problem, and until more data on the (non-nuclear) phenomenon of migration are available, care must be taken in the interpretation of the axial distribution of quantities measured nondestructively by an axial scan of the fuel. The input analysis of the reprocessing facility is of particular importance to safeguards because here accountancy changes from digital to open. It is the first point at which direct analysis of the spent fuel is possible.

The verification of the results of the reprocessing input analysis requires considerable inspection effort. Therefore the empirical finding that ratios of certain isotopes are correlated in a universal way with data characteristic of the fuel and its history has soon been recognized as a simple independent means of verification of the measured fuel composition and reported data from the reactor operator.

By far the most important, most widely-used and best studied relations are those that correlate data such as burnup, fission rates, and ratios and concentrations of fissile and fertile material with ratios of heavy isotopes /5,51-62 /. These correlations, empirical as they are, do not depend upon FPND. Only the predictions by the reactor operators do, but as FPND dependence of reactor calculations has been the subject of other review papers at this panel, this topic need not be discussed here.

Another type of correlations has also been investigated that uses fission product ratios instead of ratios of heavy isotopes. Radioactive fission products which can be measured nondestructively have been discussed in the previous chapter. Therefore only correlations with ratios of stable fission products will be considered here. Until now the following correlations have been established:

⁸⁴Kr/⁸³Kr with D5, FT; ²⁴⁰Pu/²³⁹Pu; ²⁴⁰Pu/U_{total}, ²⁴²Pu/U_{total}; ²³⁹Pu/U₀, ²⁴⁰Pu/U₀, ²⁴¹Pu/U₀, ²⁴²Pu/U₀/51.60,64.65 ]; ⁸⁶Kr/⁸³Kr with D5, F5, FT / 51 ]; ⁸⁶Kr/(⁸³Kr+⁸⁴Kr) with F9/F5 /51 ]; ¹³²Xe/¹³¹Xe with FT; ²⁴¹Pu/U_{total}; ²⁴²Pu/U_{total}; ²⁴⁰Pu/U₀, ²⁴¹Pu/U₀ ²⁴²Pu/U₀ / 63-65 ]; ¹³⁶Xe/¹³⁴Xe with ²³⁹Pu/U₀ /51 ]; ¹⁴⁶Nd/¹⁴⁵Nd with FT, ²⁴⁰Pu/U_{total}; ²⁴⁰Pu /U₀ / 5, 64, 65 ].

Here D5 is the  235 U depletion, F5, F9 and FT are the  235 U,  239 Pu and total burnup (FIMA), U₀ is the total initial quantity of uranium, and the other figures are total quantities of an element or isotope in the spent fuel.

Until now fission product correlations have been determined purely empirically. This means that although a sizeable body of data has been accumulated which is routinely used for the verification of results from new analyses, data from new dissolutions are still needed to complement existing data and prove (or disprove) the independence of a correlation of certain parameters of the fuel or its history. In order to understand the physical background of the correlations and predict their behaviour for parameter constallations for which no data have so far been available (such as plutonium-recycled LWR fuel), calculations of correlations are necessary. Predictions with existing codes have not always given good agreement with experiment. This could be due to inadequacy of the codes (such as poor choice of the neutron energy group structure), but also to lack of a sufficiently accurate set of nuclear data including FPND.

In order to investigate this latter question, a sensitivity study has been started of which first results are now available / 46 /. Table IV shows the propagation of errors in yields and capture cross sections to the computed production rate of the stable isotopes used for correlation techniques. It is obvious that most isotopes depend linearly upon their own cumulative yield.

For the krypton isotopes the situation is very simple in the case of ⁸⁶ Kr which is practically not affected by any other FPND. For ⁸³ Kr about one third of its cross section error propagates into the production rate. ³⁴ Kr is affected by 15 % of the error of the ⁸³ Kr yield and 12 % of the

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TABLE IV. SENSITIVITY OF THE COMPUTED PRODUCTION OF STABLE FISSION PRODUCTS USED IN CORRELATION TECHNIQUES (IN %) TO AN ERROR OF 1 % IN THE YIELDS AND CROSS SECTIONS OF THE FISSION PRODUCTS IN THE LEFT COLUMN, FROM  $(46, 47)^{+}$ 

contributing isotope	effect of cross yield sectio upon	n	effect yield upo	t of cross section on	e yie	ffect o ld cro ld sec upon	f oss tion
	⁸³ Kr		84 _K	ïr		⁸⁶ Kr	
81 82 Br 83 Kr 84 Kr 85 Kr 85 Kr 86 Kr	0.094 0.013 - 0.053 0.903 0.337  	, ,	0.076 - 0.148 0.840 - -	0.0009 0.005 0.121 - - -	- - - 0.0 0.9	02 0.0 95	
	¹³¹ xe		132 Xe	¹³⁴ x	e	136	Xe
129m 129 Te 129 I 130 Te 130 Te 131 Te 131 I 131 I 132 Te 132 Te 132 Te 133 m 134 Xe 134 Xe 135 Xe 136 Xe	0.0134 - 0.0448 0.0584 0.0044 0.0038 - 0.0583 0.195 - 0.737 0.0143 - 0.701        		014 - 050 0.0066 0.0065 48 - 85 0.0044 0.232 4 - 187 0.002 - - - - -	- - - - - - - - - - - - - - - - - - -	- - - - - - - - - - - - - - - - - - -	- - - - - - - - - - - - - - - - - - -	- - - - - - - - - - - - - - - - - - -
		145	Nd	14	16 _{Nd}		
$ \begin{array}{c} 141\\ 142\\ Ce\\ 142\\ Nd\\ 143\\ Ce\\ 143\\ Pr\\ 143\\ Pr\\ 143\\ Nd\\ 144\\ Ce\\ 144\\ Ce\\ 144\\ Nd\\ 145\\ Nd\\ 146\\ Nd\\ Nd \end{array} $	0.0 0.0 	0038 0023 - 49 - 13 0003 726	0.0020 0.004 - 0.0007 0.0290 0.0073 0.26 1.48	0.0006 0.0004 - 0.0042 - - 0.03 - 0.409 0.514	- 0.000 0.000 0.012 0.003 0.069 0.483 0.036	94 96 94 93 95 95 95	

t under the conditions as given at the bottom of Table III

83 Kr cross section.

¹³¹ Xe depends strongly (70 %) upon its cross section. ¹³² Xe is affected by 23 % of the yield and cross section of the mass-131 fission products. ¹³⁴ Xe is not influenced by data from any other isotopes, but for ¹³⁶ Xe the ¹³⁵ Xe yield and cross section error affect the production with 41 % and 11 %, respectively.

11 %, respectively. ¹⁴⁵Nd depends on the yield of the mass-143 chain (15 %) and yield (11 %) and cross section (26 %) of ¹⁴⁴Nd; the dependence upon the error of its own cross section is more than proportional (148 %). ¹⁴⁶Nd depends markedly upon the yield (41 %) and cross section (48 %) of ¹⁴⁵Nd and to a lesser extent on its own cross section (4 %) and on the yield (3 %) and cross section of ¹⁴³Nd (7 %).

Those effects of FPND uncertainties are rather strong. And not only do the yield values published by different evaluators vary considerably / 37, 44, 66 /, but the confidence limits of the published data are also in poor agreement. With the yield uncertainties of Ref. / 37 / (which are usually between 2 and 4 %, but occasionally as large as 10 %) and such cross section data as were available at the time of preparation of this review / 44, 66 /(between 10 and 100 % error), arithmetic addition of the components resulted in total errors from FPND uncertainties between 5 and 10 %. If one considers that cross section errors usually have the effect of giving a low value for one isotope and a high value for its parent (or daughter) and thus affect twice the ratio of the two nuclides, much of the hitherto poor agreement of calculated and measured fission product correlations can be blamed upon poor knowledge of FPND.

#### 7. CONCLUSION

The three categories of safeguards techniques that are affected by our present knowledge of FPND are quantitative assay of fresh fuel by various (mostly nondestructive)methods, the  $\gamma$  spectrometric nondestructive investigation of spent fuel, and the use of fission product correlations for verification of the result of reprocessing input analyses.

The connection of fresh fuel assay techniques to FPND is very loose because data accuracies are usually too poor for an assay method to directly rely on FPND. Instead, calibration is done by standards. For the application of these methods a number of severe problems had to be solved (neutron moderation in unknown matrix material, stability, etc.) to which FPND could not contribute. The few minor areas where knowledge of FPND (and some data usually not considered as such) can be helpful are listed in Table V. Because the return from better FPND is so hard to determine, there have until now been no sensitivity studies or the like to quantitatively establish what accuracies are needed. Also most methods now in application have to use rather coarse effects such as counting of gross delayed neutrons, gross delayed  $\gamma$  rays etc. in order to keep strengths of sources of interrogating radiation low, measuring times short, and statistical errors tolerable, and cannot afford to make use of nuclear data of single fission products <u>/ 6_7</u>.

Nondestructive  $\gamma$ -spectrometric investigations of spent fuel is interesting for safeguards if, for some reason, item accountability between the output of the fabrication and input of the reprocessing plant is impossible or has been upset and there is reason to distrust data reported by the reactor operator. In that event the measured data can be used to compute various quantities important to the safeguards authorities. Sensitivity studies as to which data are needed to improve the computation of what physical quantity pertaining to the fuel or its history are just beginning to be made; also there seems to be considerable disagreement as to which FPND can be considered how accurate. A preliminary and rather qualitative list of FPND for which an improvement of accuracy is desirable is given in Table VI. TABLE V : FPND DEPENDENCE OF VARIOUS METHODS FOR FRESH FUEL ASSAY

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priority, and reason	<pre>if low; ia- problem may be solved by empirical approach</pre>	- very low; method is not in practical use a-	new low; superiority over existing methods questionable	ut speed and accuracy of existing device is adequate for most purposes	very low; speed and accuracy ut of existing device is adequate for most purposes	ort large errors from other effects (un- čertainties about matrix material, etc.) justify simple calibration procedure
purpose	investigation o activation of c libration stan- dards	reduction of ef fort and errors associated with calibration; in vestigation of activation of c libration stan- dards	development of method	optimization of instrument layo	optimization of instrument layo	reduction of calibration eff
FPND ?	yes	yes	yes	ou	ğ	8 2
accuracy	existing data are adequate; completeness and easily use- able form are more important than accuracy	for short lived FP ratios of $2^{41}Pu/2^{39}Pu/2^{35}U$ yields for the same FP to $\pm 1$ %, energies to 0.3 keV, other data to $\pm 10^{8}$ , library of data from long-lived FP as above	Y, $T_{1/2}$ , $I_{\gamma}$ to -80/+400 %	$\frac{v_{\gamma}}{v}, \frac{v_{\gamma}(v_{\gamma}^{-1})}{v_{\gamma}(v_{\gamma}^{-1})}, \frac{v_{\gamma}(v_{\gamma}^{-1})}{v_{\gamma}(v_{\gamma}^{-2})}$ to about ±10 %	v _p (v _p -1) to about ±10 %	$\frac{v_p(v_{p-1})}{p}$ to ±1 %
data reguired	library of ²³⁹ Pu yields, half lives, $\gamma$ -ray energies and intensities for FP with half lives > 1	<pre>2³⁵U, 2³⁹Pu, 2⁴¹Pu thermal yields, half lives, Y-ray energies and intensities for most prominent Y rays from FP in the 1 second- 1 hour half-life range; library of data of FP from 2³⁵U, 2³⁹Pu, 2⁴¹Pu as above</pre>	yields, half lives, Y-ray ener- gies and intensities in the time range 1 µs to 1 s	prompt $\gamma$ -ray multiplicity from fission of ²³⁵ U, ²³⁹ Pu and their correlations with both primary and secondary particle energy	prompt neutron multiplicity distributions for ²³⁵ U, ²³⁵ U, ²³⁵ Pu, ²⁴¹ Pu	spontaneous fission multiplicity distributions for ²³⁸ Pu, ²⁴⁰ Pu, ²⁴² Pu
method	FBR assay using 252 Cf and mea- suring delayed Y rays	LWR pin assay by activation in a reactor and spectros- copy of de- layed Y rays	spectroscopy of Y rays from short-lived FP	assay of va- rious samples using ²⁵² Cf and detecting coin- cident partic- les	assay of various samples using Pu-Li neutrons and detecting coincident neu- trons	assay of plu- tonium (inclu- ding waste) by spontaneous fission neutron coincidence detection

ratios of $1^{+0}Ba^{-1}^{+}$ 1311, 141Ce, $9^{5}Zr^{-9}$ also $1^{++}Ce^{-1^{++}}Pr$ and
cf. paper no. 5 of
¹³⁴ Cs/ ¹³⁷ Cs
⁹⁵ Zr- ⁹⁵ Nb/ ¹³⁷ Cs
⁴⁴ Ce- ¹⁴⁴ Pr/ ¹⁰⁵ Ru- ¹⁰⁶ Rh ¹⁵⁴ Eu/ ¹³⁷ Cs
J.G. Tyror, Impc C. Devillers

TABLE VI. FPND REQUIREMENTS FOR NONDESTRUCTIVE INVESTIGATION OF SPENT FUEL BY GAMMA SPECTROMETRY

Correlations of ratios of stable fission products with the results of reprocessing input analyses play an important part, but are to date exclusively based upon empirical data. FPND are useful for their theoretical understanding and for predictions as to their applicability to cases for which no experimental data are now available.

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# IMPORTANCE OF FISSION PRODUCT NUCLEAR DATA FOR FUEL HANDLING

E. Merz and M. Laser

Institute of Chemical Technology Kernforschungsanlage Jülich GmbH

517 Jülich / Germany

## Abstract

In the course of reactor fuel handling, i. e., fuel storage and transport, reprocessing and refabrication as well as possible isolation of actinide elements, an accurate knowledge of the fission product compositions in spent fuel from different fuel cycles is required. Decay properties and characteristics of the respective radionuclides are also necessary. These data are needed for calculations of fuel composition, inventory and material balances, shielding requirements, decay heat development, criticality prevention, radiation decomposition of process chemicals as well as possible stored energy buildup in solid matter.

Generally speaking, the data for radioactive species of intermediate to long half-lives are known to within 5 to 20 % accuracy, and are readily available for all the various reactor and fuel types, and irradiation histories.

A survey of desirable additional nuclear data, particularly for some (n,2n)-,  $(n,\gamma)$ -,  $(\alpha,n)$ - and spontaneous fission reactions, is presented. The inauguration of an internationally accessible nuclear data library is recommended with the task of not only compiling all published data, but especially evaluating the most reliable ones. Delivery of such selected data to users in a common computer media would be additionally useful.

## 1. Specifications and technical background for required data

Handling of spent nuclear reactor fuel elements includes

- fuel storage and transport
- reprocessing and refabrication and possibly

- actinide element isolation and production processes.

All these actions require an understanding of the laws governing the formation and decay of radioactive nuclides both in the reactor and after their removal from it. This knowledge is necessary for specifying the measures to be taken during storage and transport, the separations that must be accomplished; the precautions needed to avoid criticality, the amount and kind of biological shielding that must be provided, the amount of heat that must be removed from irradiated materials, and the composition of the radioactive wastes that must be stored or otherwise disposed of.

Radioactive nuclei in spent nuclear reactor fuel emit  $\alpha$ ,  $\beta$  and  $\gamma$  radiations. Delayed neutrons are also emitted from fuel immediately after removal from the neutron flux of a reactor. But this is of minor importance on the subject discussed here. These emissions eventually end with the formation of a stable nucleus, and no further radioactivity is then released. The number and nature of the disintegrations required before the stable state is reached and the times involved are characteristics of the particular nuclide. Most radioactive nuclei go through several transformations. Precise knowledge of these characteristics is of utmost importance for the accurate and reliable calculations needed to ensure safe fuel handling.

The laws describing the physical processes are amenable to rigorous mathematical formulation. The necessary physical constants needed are:

- neutron cross sections (differential, resonance, and integral)
- fission yields (independent and cumulative yields)
   depending of incident neutron energy
- half-lives of radioactive nuclei, and

- branching ratios.

Nearly all of the data required by fuel handling users for the most important fissile and fertile nuclides  233 U,  235 U,  238 U,  239 Pu,  240 Pu,  241 Pu and  232 Th as well as for some minor important heavy nuclides are known today, though not always with high precision. Many sophisticated computer programs are available for calculating the properties of spent fuel for any conditions of irradiation and decay [1].

The following fission product nuclear data are needed.

## 1.1 Fuel composition

Concentrations, weights and radioactivities of all fission product elements must be known in order to establish the required chemical separations, procedures and to an accurate material balance. Only the average fission product element concentrations must be known for waste treatment and disposal.

## 1.2 Shielding

In fuel handling, shielding must be provided primarily to protect from  $\gamma$  emissions. Neutron shielding is necessary only for fuels containing higher quantities of transuranium elements. Precise calculational methods are available.

# 1.3 <u>Heat release during fission product decay</u>

Knowledge of the rate of energy release during the decay period is important for the design of storage, handling and transport facilities. The heat released by decaying fission products can be determined calorimetrically, or it can be calculated from the decay chains.

Radiochemical calculations suffer from uncertainties in the fundamental data of cross sections, half-lives and branching ratios.

## 1.4 Criticality

Knowledge of fission product nuclear data is of minor importance in criticality prevention since self-poisoning by fission products is not considered an adequate control in nuclear safety.

### 1.5 Radiation decomposition of process chemicals

The amount of degradation undergone by the different materials depends on the total integrated dose and, in a number of cases, also depends on the rate at which the dose is administered. Approximate values of dissipated energy are sufficient for adequately accurate estimations.

### 1.6 Stored energy buildup

When a solid is subjected to irradiation (e.g. solidified wastes containing fission products), defects are produced which cause an increase in the energy content of the crystal. This increase, called the WIGNER or stored energy, may be released by heating the material to temperatures sufficient to activate diffusion processes which allow the crystal structure to return to equilibrium state. Calculations of self-irradiation of solidified wastes by the fission products reveal that these effects do not pose serious safety problems. Estimated values of the decay characteristics of the most abundant radioisotopes are adequate for these calculations.

# 2. Assessment of present status of the nuclear data from the users view point

## 2.1 Radioactive fission products significant in fuel handling

State-of-the-art technology assumes cooling periods in the range of 120 to 200 days for spent fuel elements prior to reprocessing, actinide element isolation, refabrication, and any further treatment, in order to allow the short-lived, troublesome fission products to decay to negligible amounts, to allow certain precursors of fissile heavy isotopes to decay to fissile species, and to reduce the level of total radioactivity and decay heat.

Table I gives a survey of the relevant radioactive isotopes, including all radioactive species present in amounts

greater than about 0.2 mCi/MWd per ton of U at 120 days cooling.

Rep	rocessing Long	-Cooled Fuels	
н -3	Nb-95	I -131	Ce-144
Kr-85	Ru-103	Cs-134 ⁺	Pr-144

Rh-103m

Ru-106

Rh-106

Sb-125

I -129

Te-125m

Sr-89

Sr-90

Y -90

Y -91

Zr-95

Nb-95m

Table I:Radioactive Fission Products of Relevance toReprocessing Long-Cooled Fuels

⁺ These species are produced by neutron capture in stable fission products

Cs-137

Ba-137m

Ba-140

La-140

Ce-141

Pr-143

Nd-147

Pm-147

Sm-151

 $Eu - 154^{+}$ 

Eu-155

However shorter cooling times may be required in the future for fast-breeder reactor fuels to reduce plutonium inventory charges in the fuel cycle. Transport and reprocessing at cooling times as short as 30 days have been considered. There is thus a need for data on radioactivities of fission products which are present in significant amounts in fast reactor fuel at cooling times as short as about 30 days.

All such additional radioactive fission products are summarized in table II. These radioisotopes are generally of importance in fuel storage calculations. In addition, a few rather short-lived radioisotopes have to be included for fuel storage at the reactor site to obtain sufficiently accurate data for calculating the necessary shielding and cooling requirements (e. g. Nb-97, Ba-139, mKr-85, Ru-105 etc.).

<u>Table II:</u> Additional Radioactive Fission Products of Relevance to Reprocessing Short-Cooled (> 30 Days) Fuels

Sn-123	Te-129m	Xe-133
Sn-125	Te-129	Pm-149
Sb-127	Te-132	Sm-153
Te-127m	I -132	Eu-156
Te-127	Xe-133m	
	Sn-123 Sn-125 Sb-127 Te-127m Te-127	Sn-123Te-129mSn-125Te-129Sb-127Te-132Te-127mITe-127Xe-133m

* These species are produced by neutron capture in stable fission products.

## 2.2 Weights of fission product elements in irradiated fuels

The weights of individual fission product elements, active and inactive, are of considerable importance in the highly radioactive stages of reprocessing and in the concentration and storage of the highly radioactive wastes. It is also of value to know the mean atomic weights of these artificially produced elements. The significant fission product elements for which these data are needed are listed in table III.

~				
	H	Zr	In	Ce
	Ge	Nb	Sn	Pr
	As	Mo	Sb	Nđ
	Se	Тс	Те	Pm
	Br	Ru	I	Sm
	Kr,	Rh	Хе	Eu
	Rb	Pđ	Cs	Gđ
	Sr	Ag	Ba	Tb
	Y	Cđ	La	Dy
Ĩ				1

Table III: Fission Product Elements having Significant Weights

## 2.3 Required accuracy of the fission product nuclear data

Generally speaking, nearly all fission product nuclear data required for fuel storage and transport, reprocessing and any further treatment of the different product streams are adequately defined and known today for the various reactors and fuel types. It is sufficient, if the data for the longer lived fission products listed in table 1, are known to within an accuracy of about  $\pm 5$  to 10 %. The others need to be known only to within an accuracy of  $\pm 10$  to 20 %. However, the total decay heat should be known to within  $\pm 5$  %. Of course, it is statistically probable that this latter accuracy may be achieved even if the individual fission products are only known to poor accuracy, and this point may repay further investigation.

For example, in a recent re-evaluation on the  235 U fission product decay energy [2], summation studies made with three programs of different degrees of sophistication, using five different libraries containing different numbers of, as well as different physical parameters for  235 U fission product nuclides, indicated that the energy released is not sensitive to these differences for times > 10³ sec after fission. For practical  235 U fueled reactors, it is shown in this report [2] that neutron absorption effects on fission product energy release are unimportant, and that the  235 U fission product energy release values in the proposed ANS standard on the subject are within a few per cent of the values obtained from two recent programs and their updated libraries.

## 2.4 Availability of data

It is important from the point of view of the designers and operators of fuel handling and reprocessing plants the fission product data, as defined, are readily available for all types of fuels from the various types of reactors. That means:

-	Light water reactors	(LWR)		
-	Heavy water reactors	(HWR)		
-	Gas-cooled reactors	(AGR,	HTGR,	FBGCR)

-

Fast breeder reactors

(LMFBR, FBGCR)

- Molten salt breeder reactors (MSBR)

with their varieties of natural or enriched uranium, plutonium enriched and mixed plutonium / enriched uranium, as well as enriched uranium /  233 U and mixed thorium-enriched uranium /  233 U fuels. They must also cover the different locations of the fuels from cores, breeder or blanket zones of the various reactors. This implies that one needs to have comprehensive computer codes containing all the necessary basic nuclear data to predict the individual fission rates and, hence, the fission product radioactivities as a function of time during irradiation, for all the above variants. This in turn implies that the computer codes must correct for such things as:

- variation of neutron flux and energy spectrum during irradiation
- variation of cross sections during irradiation
- burn-out of fission products due to neutron capture
- production of active species resulting from neutron capture in stable fission products.

For any given reactor or fuel type the fission product data should cover a range of irradiation (burn-up) levels from near zero to well above the design average maximum for the fuel type, and should cover a range of fuel operating power levels extending from the lowest to the highest rated fuel in the reactor. Sufficient levels of these should be computed to allow reasonably accurate interpolations to be made.

A list of compilations, evaluations and computer codes of fission product nuclear data is presented in Paper No. 1 of this panel meeting by VALENTE [1]. It contains all important publications in this field known presently. This data library should be regularly updated and modifications periodically distributed to users.

## 3. Required additional nuclear data

3.1 The existing data compilations should be improved by measuring the energy dependent cross section functions of the following nuclear reactions in Th/U-fuel, in order to accomplish more accurate fission product (radioactivity) calculations:

²³²Th (n,2n) ²³¹Pa (n, $\gamma$ ) ²³²U (n, $\gamma$ ) ²³⁸Pu (n, $\gamma$ ) ²²⁸Th (n, $\gamma$ ) ²³³Pa (n,2n) ²³³U (n,2n).

- 3.2 The neutron emission yields from high burn-up fuels are not known well enough, in particular neutrons emitted from  $(\alpha,n)$ -reactions as well as from spontaneous fission in several transuranium elements. Much of the published data in the literature is deficient or even contradictious. Purely calculated values show considerable discrepancies with experimentally measured data. Improvement requires investigations on
  - the buildup of transuranium elements in high burn-up fuel from thermal and fast reactors (Np-237 etc.)
  - appearance of spontaneous fission and photo-fission in transuranium elements; yields and energy spectrum of emitted neutrons
  - yields and neutron spectra resulting from (a,n)reactions.
- 3.3 For the improvement of the nuclear waste management concept, that means to lower the long-term storage risk, more accurate nuclear data for the destruction of longlived α-emitting transuranium elements as well as longlived fission products by neutron capture are needed.
- 3.4 Of considerable interest and use would also be more accurate data on buildup yields of actinides under different irradiation conditions and fuel compositions (Np-237, Pu-238, Am-241, Cm-242).
- 3.5 Evaluation of existing fission product nuclear data for their reliability and accuracy to determine the best values to be used in the calculations. In case of lack of required accuracy for particular radionuclide data, remeasurement of data should be induced.

- 3.6 The form of required data, e. g. loose-leaf tables, handbooks, magnetic tapes etc., desired by the users is of lesser importance. However, it would certainly be of great help if they could be obtained from a data library in a common computer media. More important for the users however, seems to be the possibility to obtain pre-evaluated and standardized data from a data library, in order to compare calculations made at different sites. The internationally accessible library should supply their customers with a regular up-dating service for all the pertinent data mentioned in this paper (yields, decay energies, branching ratios etc.).
- 3.7 The need for more precise production rate data for lowmass nuclides, like T and C-14, important in the context of environmental hazard protection, will be discussed in Paper No. 2 of this panel meeting [3].

### 4. Conclusions

As far as fuel handling is concerned, there is no urgent need for the inauguration of a high-priority program to improve the present data status for practically all fission products described in this paper. The lack of data is illustrated in the preceding chapter 3. Most data sought in fuel handling are also of utmost importance for reactor designers, nuclear physicists and fuel cycle analysts. Their accuracy requirements are well above the ones needed for fuel handling calculations. Therefore a program for the improvement of fission product nuclear data might better be determined by these people. The technical as well as economical justification for an improvement of the hitherto available data is obvious, since the data are needed for commercial nuclear power production and utilization.

Speaking generally, the fission product data required for fuel handling and reprocessing studies are fairly modest, covering a restricted range of radioactive species of intermediate to long half-lives, and known to within 5 to 20 % accuracy. Readily available data over ranges of irradiations

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and rating for all the various reactors and fuel types are however of utmost importance.

It would be most desirable to inaugurate a world-wide compilation activity to collect and evaluate the existing (and future) published fission product nuclear data and to make them readily accessible to all users in a suitable manner, e. g. computer files etc.

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68 Mannheim, Germany

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IMPORTANCE OF FISSION PRODUCT NUCLEAR DATA IN LIFE SCIENCES, AGRICULTURE AND

#### INDUSTRIAL TECHNOLOGIES

## V.K.G. Kühn, E.-G. Niemann

Gesellschaft für Strahlen- und Umweltforschung mbH. München Institut für Strahlenbotanik Hannover, Fcd.Rep. Germany

#### ABSTRACT

In life sciences fission product nuclear data (FPND) are required for research work in human and animal sciences for the study of kinetic and static physiological processes in all organs like uptake, retention, secretion, pathways, transfer coefficients etc., and for clinical diagnosis and therapy.

In agriculture fission products (FP) are used in order to study, explain, and attempt to influence and control various biological, physical and chemical processes occuring in plants, food and soil in relationship to soil-plantnutrient and soil-plant-atmosphere-irrigation studies. Furthermore, experiments on cultivation, mechanisation and translocation are carried out with different FP.

In various industrial technologies FPND are required for research work and for development of equipment for measurements of thickness, density (mass per unit area), coating thickness, moisture and pressure, as well as for mining, level gauging, charge elimination, luminous devices, energy sources and for gamma radiography.

These investigations with FP as tracers, consisting of different radiochemical compounds, and the applications of very different radiation sources for industrial, medical, agricultural and laboratory use require the knowledge of the following fission product nuclear data and other information:

- 1. half-life
- 2. type of radiation
- 3. decay-schemes including all excited states and transitions to the ground state level energies and life-times, types of radiation and percentages per decay.
- 4. purity and data of impurities
- 5. specific activity
- 6. production processes other than fission (including the production crosssections)

- 7. dose-constants for gamma-radiation
- 8. Auger electrons
- 9. beta spectra a) pure
  - b) changed by capsule-material
- 10. calibration accuracy
- ll. chemical compounds: solubility, pH-value, costs.
- 12. concentration of stable nuclides present in sources of mixed FP
- 13. Bremsstrahlung sources: efficiency, photons/beta; K-X-ray production of the target.

#### 1. INTRODUCTION

Before any real progress could be achieved in the application of radioactive material, which is available since the first year of the twentieth century, it was definitely necessary to measure and to present data and other information on the radiation emitted by the radioiscopes used. Since the time when the first nuclear reactors came into operation, fission products became available to users and nearly all fundamental data of the nuclei formed by the fission of heavy elements were collected and published by numerous institutions.

Cnly about 10 % of all the FP that can be found in compilations are actually used in life sciences, agriculture and industrial technologies. However, about 25 % of the longer-lived fission products, which are produced in thermal-neutronfission with a yield of more than  $10^{-9}$ % from U-235, U-233 and Pu-239, are really useful in the fields discussed here /10/. Although they represent only about 2 % of all known radionuclides an astonishing high part of FP are applied. From this point of view it is very valuable and important to revise FPND from time to time and to explore whether and which additional information is required.

#### 2. TECHNICAL BACKGROUND FOR THE APPLICATION OF FP.

### 2.1. Life sciences

FP in human and animal science provide a means to study the chemical situation of the body by labelling most of the constituents, and to follow them through bio-chemical and physical processes. Research work in this field can be specified as studies of static and kinetic physiological processes in all organs concerning the metabolism like uptake, retention, secretion, pathways, transfer coefficients of elements and compounds as well as the localization of pathological changes.

When using radicactive material as a tracer in life science the user is responsible for minimizing harmful effects. Therefore activities can only be introduced into the body in very small quantities for tracer work. For this reason the quantities and the activities required are generally relatively small and the accuracy of the date involved has to be relatively high.

Typical applications of FP are clinical function tests with tracers for Thyroid gland, diagnosis, treatment and research in hematology, location of brain tumors, plasma and blood determination, plasmaprotein-antibody metabolism, szintigraphy, whole body metabolism, circulation and flow measurements etc. /1,8,12,13/.

Apart from these studies in physiological and pathological research, FP are used as intra- and extracorporal radiation sources for therapy and for diagnostic radiography including dental radiography. Some of the FP have been found suitable for therapy by local treatment as enclosed radiation sources, useful for completion and for investigation of new possibilities in radiology.

Without demanding a complete enumeration it is obvious that a tremendous field of applications of FP is involved in medical research, diagnosis and therapy. A good part of the progress which could be attributed to classical medical research work is based on the availability of the following FP: H-3, H-3,

Ga-72, Br-82, Kr-85, Sr-89, Sr-90/Y-90, Y-91, 2r-95/Mb-95,  $Mo-99/Tc^{m}-99$ , Ru-106/Rh-106, Ag-111, I-129, I-131, I-133, Te-132, Xe-133, Xe-135, Cs-137/Ba-137^m, Ba-140, Ce-144/Pr-144, Pm-147, Sm-155 and Eu-155. For biological aspects of the environmental contamination produced by man (nuclear power stations, nuclear weapons), the generation of FP in fission has to be taken into account additionally. This means that FP activities at various times after fission and the nuclear data of the FP involved have to be known, at least for those FP which contribute more than 5 % of the total activity at any time /5/.

## 2.2. Agriculture

Research work with radioisotopes as tracers in agriculture, in laboratory as well as in field experiments has resulted in the improvement of the use of fertilizers. This led to a better understanding of soil plant relationship, to a significant increase of yields, and to new cultivation methods /2,4,6,16/.

Fost of these experiments are carried out with radioisotopes of the same element and these radioisotopes are only in very few cases FP. As far as FP are involved in agricultural research they are used:

- a) as irradiation sources
- b) as tracers for translocation and penetration investigations in connection with fall-out-studies as well as with soil-plant-nutrient research and other agricultural problems;
- c) for density-thickness and moisture measurements (mass per unit area) in biological substances and soil. (see "Industrial Technologies")

For acute and chronic irradiation to study dose- and dose-rate-effects on different species of higher plants mainly Cs-137/Ba-137^m is used as radiation source. The effects produced include morphological and histological changes, mutations, and growth stimulation. The research fields concerned are plant breeding, radiation cytology and genetic studies, chromosome and cellular radiosensitivity investigations and radiological survival studies. Sterilisation of soil, food, insects (sterile male technique) and also sludge is an other important field of FP application.

For the purpose of chronic irradiation in genetical, botanical and ecological research in connection with fall-out studies, so called fallout-decay-simulators of several 100 to nearly 10.000 Ci Cs-137 are applied /14/. Also mixed FP (different ages from 10 days to 10 years with various percentages of the components) or isolated FP like I-131, Ce-144/Pr-144, Ru-106/Rh-106, Ru-103, H-3, Cs-134, Cs-137, Sr-90/Y-90, Sr-89 and 2r-95/Nb-95 are applied for the determination of deposition, transfer and accumulation of those materials in ecosystems as functions of climatological, physiological and soil parameters /15/.

In soil-plant-nutrient experiments Rb-36 seems to be extremely suitable for studies of root system development of plants (esp. fruit-trees) and Sr-89 as tracer for calcium uptake in plants. Furthermore, Sm-153; Ba-140/La-140 and Zr-95/Nb-95 are used for labelling of sand.

# 2.3. Industrial Technologies

FP as radioactive tracers as well as the application of encapsuled fission products as radiation sources have contributed to the solution and understanding of many industrial research problems /3/. Apart from the studies in industrial laboratories a great number of equipment in raw material industry and in many other factories is furnished, using radioisotopes to precisely measure and to control production processes. New and improved methods of production are the result of the use of ionizing radiation in non-destructive-test procedures. A lot of technological problems connected with automatic operations could be solved with FP. Profitable use was made in the production of basic metals, chemicals and plastics, paper, petroleum, rubber, textile, in coal-mining, food, machinery, minerals, buildingtrade, subsoil and many other industrial branches. Energy production by isotope powered generators is gaining increased importance /9,11/.

FP used in industrial technologies and their specific applications are listed below in detail:

Br-82:	Tracer (leak-search); mass per unit area;
Kr-85:	Luminous devices; Bremsstrahlung-sources (absorbed in active charcoal); ventilation; static charge eliminators;
Sr-89:	Tracer; calibration of instruments;
Sr-90/Y-90:	Tracer; mass per unit area, charge eliminator; thermoelectric generator; level gauging, Bremsstrahlungs- sources (sandwiches between Al-foils or in Pb-matrix);
Y-91:	Mass per unit area;
Zr-95/Nb-95:	Tracer (sand); instrument calibration;
Ce-144/Pr-144:	Mass per unit area; radiography; thermoelectric generators;
Ru-106/Rh-106:	Mass per unit area; Bremsstrahlung-sources (various targets).
I - 131:	Tracer (water and others)
Te- 132:	Parent of I-132
I <b>-</b> 132:	Tracer (water)
Cs- 137/Ba-137 ^m :	Irradiation sources; mass per unit area; radiography; density - logging; level gauging;
La- 140:	Flow - motion of concrete
Pm- 147:	Mass per unit area; self-luminous paints, Bremsstrahlung sources (Ag-matrix, Al-matrix) thermoelectric generators; radiography;
Eu - 155:	Radiography

Measurement of mass per unit area can be calibrated as determination of density, thickness or moisture respectively.

## 3. TYPES OF FPND AND OTHER INFORMATION REQUIRED

Data that are generally required for the applications of FP in the above mentioned fields are:

l. half - life;

- 2. type of radiation;
- 3. energy;
- 4. decay schemes (including level energies and life-times, types of radiation and percentages per decay);

- 5. cross-sections in the thermal and fast neutron energy range;
- 6. production processes other than fission; other isotopes produced in these processes;
- 7. dose-constants for gamma radiation;
- 8. Auger electron production;
- 9. beta spectra a) pure

b) changed by capsule - material

- 10. purity and data of impurities (for example the fraction of Cs-134 in a Cs-137 source at any time);
- 11. specific activity;
- 12. calibration accuracy;
- 13. chemical-compound, pH-value, sclubility, costs;
- 14. concentration of stable nuclides present in a mixed FP source.
- 15. Bremsstrahlung sources: efficiency, photons/beta; K-X-ray production of the target.

This enumeration involves a number of additional data which are not given in former lists but which are extremely important for special applications /7/. Revised data should be given and more exact data should be available for:

energies:

specific activity;

Auger-electrons;

purity and impurities of longer-lived isotopes (for example: if Sr-89 is used, knowledge of the Sr-90 contamination level is necessary);

nuclear data of parent and daughter, also in cases where only the daughter product is used.

precise knowledge of the abundances of radionuclides present in a fission product sample; changes of Beta-spectra by capsules.

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#### USE OF FISSION PRODUCT NUCLEAR DATA IN LIFE SCIENCES

Edward L. Alpen, Director Pacific Northwest Laboratories Battelle Memorial Institute Richland, Washington

#### Abstract

This paper describes fission product nuclear data as it applies to the life sciences in seven situations. They are: external exposure; internal emitters; medical application; sealed sources for Brachy therapy; sealed sources used in teletherapy; environmental pathways; and isotopic applications. Sufficient data are available for the applications discussed.

I will identify seven general situations for application of fission product nuclear data and of data on the fission process in uranium and transuranic elements:

- Toxicity to exposed individuals from externally located isotopes, i.e., isotopes in the environment or in the fuel cycle.
- Toxicity caused by deposition of radioactive materials in living systems.
- Medical application of injected radioisotopes for diagnostic or therapeutic purposes.
- Use of sealed sources for implantation in living tissue. (Brachy therapy)
- 5. Use of sealed sources at a distance from the patient for therapeutic purposes. (Teletherapy)
- 6. Environmental pathways for uncontrolled release of materials into biosphere.
- And, finally, isotopic applications in medicine, biology, agriculture, and industry including gamma ray and neutron radiography.

1. External Exposure. Exposure of human beings to ionizing radiation sources may occur either accidentally by loss of process control or accidental release into uncontrolled environments, or it may occur under controlled conditions with pre-established exposure limits. Typically, the former may occur with effluents from large production and processing facilities for nuclear fuels.

Almost without exception these exposures occur as the result of releases of mixed fission products and activation products of complex age and history. For this reason exposure control and dose estimation are made on the basis of total ionization measurements, and nuclear data are needed only for prediction, regional modeling for future events, and for shielding calculations. Rarely is exposure to beta rays a significant hazard under these conditions, and only gamma ray data on fission yield and halflife are essential.

The same generalizations may also be made for exposure under controlled conditions, but here beta dose is often important for exposure of extremities, and beta ray spectrum is useful for dose prediction models.

In any of these cases half-life data for short-lived nuclides is generally not significant. By short, I would indicate that nuclides with half-lives of several hours or less are of little importance.

An unusual but important case in the external exposure category is that of criticality accidents. Since dosimetry must usually be reconstructed, it is essential to know the prompt neutron spectra--both thermal and fast, the prompt gamma spectrum, and the ratio of each.

Data are generally adequate for all of these applications, and accuracy in the 25-40% range is adequate.

2. <u>Exposure from Internal Emitters</u>. Internal emitters constitute a most significant source for human exposure to both fission products and transuranic elements.

The dosimetry relating to estimating hazards from ingested radioactive materials is extremely complex and troublesome. The

material is rarely, if ever, uniformly distributed throughout the body of the exposed person; in fact, it is rarely uniformly distributed in even the individual organ of interest, such as bone and bone marrow for Sr-90 and Cs-137 or lung and lymph nodes for plutonium isotopes.

The data that must be known to derive a dose estimate for an organ are the concentration, of course; the number of disintegrations per unit time, and the average energy deposited in the volume of interest per disintegration. All but the first are derived from nuclear data, but other considerations are equally important.

The concept of "effective half-life" has been derived as a useful tool to assist in this dosimetric problem. It is calculated as a resultant half-life taking into account both radioactive decay and excretion from the body. It may also be quite different for different tissues of the body, and even for different regions of the same tissue.

Given these generalizations, it is clear that we need to know the physical parameters of the decay process, but it is also clear that we rarely need to know them with high precision. From the scientific view we are satisfied with the present state of knowledge of fission product nuclear data, and from the radiation protection point of view the data are more than adequate.

The data which are presently limiting our capabilities to deal with deposited radionuclides are generally chemical data, i.e., solubilities, chemical form, etc., particularly for plutonium and other transuranics.

3. <u>Medical Application</u>. When radioisotopes are used by injection for diagnostic or therapeutic purposes, we generally need the same data mentioned before for internally deposited nuclides, since either protection of the patient or calculation of the therapeutic radiation dose to the organ of interest is the objective.

For the special case that the isotope is being used for visualization of an organ by means of scintillation cameras, we need to know the gamma energies and the disintegration rates with reasonable precision. Again, biological processes are such that the precision with which they are known is generally more limiting than the present precision of physical measurements.

4. <u>Sealed Sources for Brachytherapy</u>. A number of radionuclides are in use to supplant <u>radium</u>, usually for economic reasons, but also occasionally because they are medically superior. For examples, isotopes of iridium, gold, and tantalum are in present use.

Obviously, we need to know with some precision the nuclear properties of these materials, particularly the gamma and beta energies as well as the decay rate. Since the sources are usually encapsulated, empirical data are usually derived by ionization chamber measurements to estimate dose. A new and unusual brachytherapy source is now coming into use, spontaneously fissioning Cf-252. Data are still being completed on estimation of the dose from this source, and because of the unique difficulties associated with neutron dosimetry, it is important to have as complete data as possible on neutron spectra. The neutron spectrum of Cf-252 is already well known, however.

5. <u>Sealed Sources Used in Teletherapy</u>. Only two nuclides have had wide usage; these are Co-60 and Cs-137. For dosimetry of this class of sources we need to know and do know the gamma ray energies. For design of sources we need to know specific decay rates. Half-life is not important since only radionuclides with half-lives measured in years are useful.

6. <u>Environmental Pathways</u>. We are rarely, if ever, interested in dose delivered to plants or even animals from environmental sources. We need to know ultimately the dose to man as the result of ingestion of foods prepared from plant and animal sources. We therefore generally need only to know how to identify and measure radionuclides incorporated in food chain members.

7. <u>Isotopic Applications</u>. There are usually very convenient ways to bypass the need for precise physical knowledge about a radionuclide being used as an isotopic label. Use of decay standards is routine and eliminates the need for accurate half-life data. The energies associated with the decay need only be known in general. An exception, of course, is the need for very accurate gamma ray energy data for gamma ray spectrometry of mixed trace radionuclides, either fission products or activation products.

Summary. Nuclear data within the charter of the IAEA Panel on Fission Product Nuclear Data appear to be more than adequate for the above user applications.

# STATUS OF NEUTRON CROSS-SECTIONS OF FISSION PRODUCT NUCLIDES

#### P.RIBON and J.KREBS

# Department of Reactor Physics and Applied Mathematics Centre d'études nucléaires de Saclay, France.

### SUMMARY

The libraries of nuclear data on fission products are expected to cover some 600 to 800 F.P.'s, but the number for which a certain precision is required on the neutron reaction cross-sections is less than 150. In order to clarify the needs we propose a classification of these F.P.'s according to the absolute precision necessary.

Nearly all the requirements are for capture crosssections. A comparison of experimental or evaluated values with these precisions shows :

- that in the thermal range our knowledge of  $\sigma_e$  must be improved for a dozen nuclei (table IV), while large disagreements exist for some thirty others.
- that the R I value must be improved for about ten nuclei (table V), this value being little known for ten or so others.
- that the accuracy on g in the fast neutron field <u>must</u> be increased for some thirty nuclei, generally stable or longlived (table VI).

Comparing the values of the parameters used for calculations with the statistical model we observe very large divergences not only for parameters such as "a", which depend on the model (table VIII), but also for parameters directly deduced from experimental results, such as Dobs (table VII).

#### INTRODUCTION

The latest nuclear data libraries on fission products (F.P.) list about 600 isotopes in their fundamental or in metastable states ; the E N D F system is expected to include 800 in its 5th version. It is almost impossible to draw up an exhaustive survey of known facts concerning the neutron crosssections of these nuclei, but the task is simplified by the lack of experimental information on all but about a third, added to which they are not all equally "important" and the same attention need not be paid to each.

In principle the estimation of the importance of an F.P. and of the accuracy required on the cross-sections is outside the scope of this review. However it is clearly impossible to consider 150 to 250 nuclei without some guiding principles, bearing in mind especially that the precision required on a given datum varies considerably from one nucleus to another and that certain data are seldom or never requested.

In the first section of this report the idea of importance will be examined and a classification proposed whereby the absolute precision required on a given datum is the same for all nuclei of the same group. With the help of explicit requests for data, mainly expressed in WRENDA, average accuracies applying to the capture cross-section for each group of this classification will be defined.

#### . FISSION PRODUCT CLASSIFICATION

## 1.1. According to "importance".

1.1.1. Criteria adopted.

The importance of an F.P. depends on the application foreseen. It will not be the same for a thermal as for a fast

reactor, will depend on flux, fuel cycle period, etc..., while there can always be special reasons to take a particular interest in one nucleus or another.

Many classifications have been proposed, some of which are given in tables II.1 to II.8. For the purposes of this study a system based only on half-life and yield is suggested.

<u>Yield</u> : The value adopted is the highest of the four yields following :

- yields from thermal neutron induced fission of  233  U and  $^{235}_{\rm U.}$
- yields from fast neutron ( ~ 2 MeV) induced fission of  239 U and  238 U.

The values are those given in the table of MEEK and RIDER [Me 72].

<u>Half-life and equivalent yield</u> : The shorter the half-life, the less important the fission product. The importance of an F.P. in a reactor will be proportional to the product of the yield by the average time it remains in the reactor. The in-pile time will depend both on the half-life of the F.P. and on the fuel irradiation period. Two irradiation times have been considered : 3 months and 1 year.

This amounts to saying that at equal capture crosssections two F.P.'s will have the same importance if the mean number of nuclei present in the reactor is the same (averaged over the fuel irradiation period), allowing for radioactive descendants but neglecting evolution by radiative capture. The "equivalent yield" is the yield of a stable F.P. with no long lived parent to prevent its formation and having the same importance, i.e. the same average time of presence in the reactor.

Under these conditions (i.e. neglecting absorption)

# the needed absolute accuracy on a nuclear datum will be inversely proportional to the equivalent yield.

Table I shows the equivalent yields adopted.

## TABLE I

Equivalent yields and absolute accuracy necessary

(order of magnitude)

	Equivalent yield	Abs	olute accur	acy needed	on
Group	ક	о ^г с 2200 m/s	RI C	с с 30 KeV	FSA*
1	5	5 b	25 b	25 mb	5 mb
2	2	10	50	50	10
3	1.0	25	100	100	25
4	0.5	50	250	250	50
5	0.20	100	500	500	100
6	0.1	250	1000	1000	250
7	0.05	500	2500	2500	500

*F S A : Fission spectrum average

1.1.2. Criticism of this classification.

It accounts for only one aspect of the problem posed by F.P.'s: their evolution in a reactor and the effects on this reactor. It also ignores absorption, which is responsible for:

 a) - The appearance of new nuclei formed by neutron capture ; this applies especially to thermal reactors, and examples will be given of cases where large quantities of nuclei are formed by this process ; b) - The saturation phenomenon : when the capture cross-section is very large (  $\sigma \phi \gg \lambda$ ) the capture rate depends no longer on the cross-section but only on the yield. This is the case particularly for ¹³⁵ Xe, to which we shall return later.

In an 3th group we have listed nuclei which are not included strictly in our 7 groups but are taken into consideration by other authors. These are nuclei :

- with equivalent yields just below 0.05 (limit of the 7th group; this applies to ¹⁰⁵Ru, ¹⁵¹Pm, ¹³⁴Cs, ¹⁴⁸Pm,...)
- or involved in other applications (astrophysics :  96 Mo and  110 Cd).

A few nuclei of marginal importance remain (⁸⁷Kr, ¹⁰⁰Ru, ...)

# 1.1.3. Comparison of different classifications according to importance.

Tables II show the results of several other classifications. Taking into account the cross-section values these classifications are consistent with ours.

In the case of thermal reactors for instance the nuclei taken as important are those having :

 $\sigma_{c} \ge 2$  b. for group 1  $\sigma_{c} \ge 10$  b. for group 2  $\sigma_{c} \ge 200$  b. for group 5

There are some exceptions : in group 2 the category "F.P.'s important for thermal reactors" includes  95 Zr but not  107 Pd or  108 Pd ; similarly the presence of  131 I (gr.4) and  135 I (gr.7) in this category may seen debatable.

#### 1.2. Other classifications.

Other classifications may be imagined, such as that proposed by the Japanese [Sa 73]: F.P.'s are divided into 3 groups according to whether or not saturation is obtained, with the exception of a few chains (A=135,149,...) considered important and dealt with separately.

## 2. PRECISION REQUIRED ON DATA.

Table II lists the requests for cross-sections according to WRENDA 73 [Wr 73], supplemented as the case may be by other demands [Us 73, Fr 73]. <u>Almost all requests are for cap-</u> ture cross-sections.

## Table II: CLASSIFICATIONS, GROUPS 1 - 8

Contents of Table II:

Classification	of fission	products	according	to	importance
				50 80m etti -	

Thermal reactors	:	J =	Sa 73
	:	¥ =	W.H. Walker - Us 70 - Sa 73
	:	B ==	BNL list (1971)
Fast reactors	;	J ≂	Ja 73
	:	U =	Us 70
	:	C ≖	Cadarache (France) - Fr 73
All reactors	:	E3=	ENDF/B3 - Li 71, Eb 73 & E4 = ENDF/B4

Evaluated Data :

- (

	:	E	1	ENDF/B3
i.	;	C	101	Australian library
	:	B	3	Italian library & B2 = capture data only
	:	$\mathbf{L}$	*2	Dutch group cross section library

	and a part of the second second	Marinum	Ord	Order of magnitude					Importance							WRENDA Requests									a
Isotope	Half- life	yield	Jc (2200)	RIc	c(30keV)	FSA	Th re	eri ac	tal	Fa re	st act	or	<b>a</b> 11	Therm	<b>a</b> l	1/	E	Fas	t	Res.	р.		Dat	ta 	
-Manual Ale dide TES (Tile		%	barns	barns	mb.	gb.	J	¥	8	3	U	c		8	P	*	P	8	ρ	3	Ρ	E	с ғ	) L	
885R	STABLE	5.6(13)	<0.1	<0.1			-					-							[]		Π		C 8	5 L	
905R	28.9 Y	6.5(03)	1.		20.	3.	[			J		i	E4	1									Ċ	L	
92ZR	STABLE	6.4113)	0.3	0.6	40.	7.	(			-			84				11						6 8	L	
93ZR	9.5858	6.8(13)	1.5	(10.)	100.	15.	]		32	3			E4	}			]	10	2				C 8	11 L	
94ZR	STABLE	6.5(13)	<0.1	0.3	20.	6.	[					ļ	84										CF	3 L	
96ZR	STABLE	6,3(U5)	0.1	6.	30.	10.	ł						84										CE	s t.	
97#0	STABLE	6.0(U8)	2.	15.	400.	40.	J		30	3	13	С.	E3			10	2	10	2			ε	C 8	3 L	(2)
98M0	STABLE	6.0(18)	0.15	7.	100.	40.	[				23	C						30	2	Į		E	0.8	) L	
99TC	2.1E5Y	6.4(U8)	20.	300.	900.	70.	J	13	13	J	2	C	E 3	5	1	20	2	10	2	10	2	8	6 8	31 L	(2)
100#0	STABLE	6.5(PU9)	0.3	5.	70.	16.					22	C										Ε	6 5	I L	1
101RU	STABLE	6.6(PU9)	4.	80,	1000.	100.	J		28	J	4	С	83					10	Z	10	2	ε	6 8	3 L	
102R U	STABLE	6.7(PU9)	1.4	7.	300.	. 08				J		C	E 3					10	2	10	2	3	C 8	3 L	
104RU	STABLE	6.51PU91	0.5	5.	160.	30.				J	1	C	(					10	2	10	2	ε	CE	3 L	
105PD	STABLE	5.1(PU9)	12.	80.	1200.	120.	J	ы		J	3	C	83			10	2	10	2			8	C 8	5 E	
132XE	STABLE	5.4(PU9)	0.3	(1.)	80.	(15.)					26												0.8	I L	(1)
13355	STABLE	6.8(PU9)	30.	400 s	650.	60.	J	9	10	J	7	С	E 3	10	11	10	11	10	1	}		ε	C 2	11 L	12)
134XE	STABLE	7.5(08)	0.25	(2.)	(30.)	(10.)																	C 8	1 L	1
135C S	2.3664	7.4(PU9)	9.	60.	(200.)	(15.)		쓝	29	J	16	C	E 3			10	2	10	2	10	2	ε	C P	1 L	1
136XE	STABLE	6.9(03)	0.25	0.3	6.	(3.)																	C 8	L	(1)
13765	30.2 7	6.5(PU9)	0.1	0.2	20.	2.				J												Ε	Cε	1 L	1
138BA	STABLE	6.7(05)	0.4	0.2	(4.)																		C 8	i L	1
139LA	STABLE	6.8(PU9)	9.	14.	50.	6.	J	M	19			С	E 3			10	2	-		10	2	E	C 2	1 6	
140CE	STABLE	6.4(13)	0.6	0.4	25.		[											i	11				C 8	i L	
142CE	5.E16Y	6.6(15)	0.9	1.	50.	10.	1																C 8	5 L	1
143ND	STABLE	6.0(05)	330.	100.	300.	30.	J	2	3	J	17	c	٤3	10	1	10	1	10	2			E	C 8	i L	

TABLE II - GROUP 1

(1) - High production by thermal neutrons capture (2) - Request for  $\sigma_{in}$  at 50% accuracy [Pr 73]

		Moziaum	Ord	er of mag	znitude			I	apo	rta	inc	e		١	RE	NDA	r	eque	st	8		Ev	al	uat	ed	
Isotope	Half- life	yield	ন্ত(2200)	RIc	d(30kev)	FSA	Th re:	erm	el or	) rea	Fas act	t or	A11	Therms	1]	1/E		Fas	t	Res.	p.		da	ta		
		%	barns	barns	mb.	ab.	1	W	8	J	IJ	с		2	P	8	Ρ	8	ρ	X	p	£	c	B	L	
BOKR	STABLE	2.8 ( 13)	0.6	<0.1	10.	2.												7		[			C	8	L	
87RB	5.E5Y	4.0 ( 13)	0.1	2.5	30.	з.														Ì			C	8	L	. 1
895R	50.8 D	5.8 ( U3)	0.5	0.8	30.	2.	1																C			
85 Y	STABLE	5.8 ( 03)	1.3	0.5	30.	3.														l			C	82	L	
91 Y	58. 8 D	5.8 ( U5)	1.2	1.4	40。	6.												1					0			, <u></u>
9128	STABLE	5.8 (U 5)	1.6	7.	100.	10.																	C	8	L	
95ZR	65.5 D	6.5 ( 15)	(2.)	7.	180.	10.		Ħ					E 3	20b	2	50ъ	2					Ε	C			1
9580	STABLE	6.5 ( U5)	140	105.	400.	40.		20	24	1	12	( C	E 3			10	2	10	2	l		E	C	B	L	(1)
10380	35.8 0	6.5 (PU9)	6.	(10.)	800.	50.		¥				C	E 3	106	2	506	2					E	C			
103RH	STABLE	6.5 (PU9)	150.	1050.	1000.	90.	J	5	8	J	5	С	83	150	2 ]1	Oup	2	20	1			Ε	C	8	1.	(1)
106RU	368. D	4.5 (PU9)	0.14	1.5	100.	20.	1			J			E 3				ļ		ĺ	Ì		ε	C		L	
10790	7.68	3.6 (PU9)	10.	70,	1100.	100.				J	8	C		10	2	10	2	10	2	10	2	E	C	81	L	
10890	STABLE	2.6 1PU91	11.	230 e	250 e	20.					19	C						10	3	10	2		C	8	L	
131XE	STABLE	4.2 (PU9)	100.	850.	500.	40.	J	7	7	J	18		£3	10	2	10	2	10	2 ;	10	2	٤	C	8	L	
14108	52.5 0	6.1 (PU9)	29.	27.		13.	J	¥					E 3			1			ł			E	C			
141PR	STABLE	6.1 (PU9)	12.	18.	150.	18.	J		20		20	C	E 3			10	2	10	2			5	C	8	L	
144CE	284. D	5.2 ( U5)	1.	2.5	50.	8.				J					ł							1	C			
145ND	STABLE	4.0 ( 15)	50.	280.	400.	30.		12	9	3	24	C	E 3	10	1	10	1	10	2	10	2	8	C	8	L	
146ND	STABLE	3.4 (U 8)	(10.)	(3.)	100.	20.	3		26									10	2				C	8	L	

TABLE II - GROUP 2

(1) - Request for  $\sigma_{in}$  at 50% accuracy [Fr 73]

TABLE II - GROUP 3

	Welfe	Novinia	Or	der of r	nagnitude			1	про	rt	anc	e		,	WR I	ENDA	re	ques	ste	3		Ev	al	uate	a	
Isotope	life	yield	Jc(2200)	RIc	(JOkeV)	FSA	Th re	er: act	al or	re	Pas act	t or	All	Therm	81	1/1	E	Fast	t	Res.	.p		da	ta		
		%	barns	barns	mb.	mb.	J	¥	8	J	U	c		:	ρ	¥	P	*	P	8	0	ε	с	8 1		
83KR	STABLE	1.01(U 3)	200.	230 .	(400.)	40 .		17	18				E3	10	2	10	2				Π	E	C	B	. 1	1)
85R8	STABLE	2.21( U3)	0.6	5 * 25	300.	25.		ы					53	1.65	,							e	ç	BI	-	
10 SAG	STABLE	1.6 (PU9)	90.	1450.	1500.	100.		18		J	9	с	E 3	10	2	10	2	10	þ			E	č	B1	-	
1281E 129 I	1.6E7Y	1.69(U 3)	30.	30.	40. 500.	50.	J			J	25	c											C	82 1 81 1	-	1
1301E 1408A	STABLE 12.8 D	2.3 (U 3) 6.3(U 5)	0.2	(1.)	10.	3. 15.	J																C C	82 1	-	
143PR 144ND	13.6 D 1.E15Y	6.0 (U 3) 5.2 (U 5)	100. 4.	170. 5.	100.	40. 20.	1	¥	22	J			E 3	5	1	20	1					8	C C	B   I		1
147PM 148ND	2.E Y STABLE	2.6 (U 8) 2.1(U 8)	170. 2.6	2200. 15.	900.	120. 25.	J	4	6	J	6 29	С	E3	10	1	10	1	10 20	2 2	10	2	F	C C	B1 1 B 1	•	2)
1495M 150ND	1.E15Y STABLE	1.84(U 8) 1.29(U 8)	40000. 1.5	3300 . 8 .	1400.	150. 40.		3	2	J	11 28	С		10	2	10	2	10	2	10	2	ε	с с	B   L B   L		3)

(1) - Request for  $\sigma_{t}$ , E(1keV, 10% accuracy, priority 2 (2) - Request for  $\sigma_{t}$ (3) - Saturation effect for  $\phi_{th} = 3.10^{19} \text{ cm}^{-2}$ 

TABLE II - GROUP 4

			Order	of magn.	ltude	nar de de ar or or o		Im	por	tar	ice	8	-	WR	EN	DA Req	ues	ts	ago (1974) Alem 1	-	Eve	lua	ted	
Isotope	Half-	yield	o _c (2200)	RIc	C(30keV)	FSA	TI	ner eac	nal tor	rea	as ct	ţ or	A 11	Therm	al	1/2	Fe	ist	Res.	p	I	)ats		
	1110	Æ	barns	barns	mb.	mb.	J	¥	8	J	U	c		Z	P	t P	*	P	3	р	8	C B	L	
82 SE 9 SMO 106PD	STABLE 67. H STABLE	0.70(U 3) 6.4 (U 8) 4.5 (P(9)	(1.) 3. 0.3	(0.4) 25. 8-	70.	10. 20.	_				21	c	F3	166	2	5002					E	B C C B	1	
11 OPD 127 I	STA2L5	0.79(PU9) 1.07(U 8)	0.25	(10.)	150. 700.	20.		•1			2.	c										C 8 C 81	L	
1327E	8.1 D 78. H	4.2 (PU9) 5.3 (U 8) 6.8 (PH9)	<0.9 <0.1	<0.1 (300.)		20. 0.2	.1	9 2	23				E 3	250b 5	2	300002					E ( F (			
147NO 151SM	11.1 0 93. ¥	2.6 (U 8) 0.93(U 8)	50. 15000.	650. 3000.	20 00 .	50. 300.	J	W E	4	J	10	с	E 3 E 3	10 5	i	10 1 10 1	10	2	10	2	E	C C 81	L.	(1)
1525M	STABLE	0.6719491	200.	3000.	500.	70.		10	17				E 3	5	1	102	1(	<u> </u> 2	10	2	8 (	<u>с</u> в	٤ 	

(1) - Saturation effect for  $\varphi_{th} \equiv T \sim 7.10^{19}$  cm

TABLE	8 4	1 .17	and a state and the															and an effect of	-						50a /1		
		Kazzme.	01	.in 60	×″ ~		1		ì	τ3.	nc.			۴	٠,	114	ЯF	ų	- - F - 3				-	n -1 -5	9.2	)	\$
Laotope	Valr- life	yrold	G(2200)		Sel 232.		,	78A	584 L 6750	(	FE: Pot	¥ 4 Z	iš.	Thole	٤.	1/8		Fas	, t	Res	. p	\$ *	D	879 8	.	) •	
		1 pr	bergs	barns	Eò.	mb.		B	B	J	υ	c		8	٩	×.	ρ	2	P	8	P	E	c	8			1
805E 818R 85KR 105RH 111CO 1265N	STABLE STABLE 10.8 Y 35.5 H STABLE 1.E5 Y	0.25(U 3) 0.36(U 3) 0.49(U 3) 5.1 (PU9) 0.37(PU9) 0.28(PU9)	0.6 3. 8. 17000. 23. 0.3	1. 50. 9. 15000. 50. 0.2	30 • 400 • 180 • 700 •	5. 40. 15. 40. 50. 4.	33	3	12			c	E 3	5	1	20	~					E	000000	6 6	22	<b>31)</b>	
129764 1430 E 14784 140PN 153EU 1548M 155EU	340 D 330 H 10E11V 5301 H STABLE STABLE 50 V	0.33(PU9) 6.0 (U 3) 2.6 (U 8) 1.84(U 8) 0.48(PU9) 0.32(PU9) 0.26(PU9)	0.4 5. 70. 1600. 420. 5. 4000.	7. (10.) 600. 800. 1500. 30. 2000.	1100. 2500. 400. 2000.	10. 10. 120. 70. 250. 50. 400.	J	W 14 14 16	31	t t	15		Е Э Е З	1000k 1002 2 10	3	1000b 5 10	2 2 2	20 10 10	5 5 1	10 10 10	2 2 2 2 2 2	e a a	0000000	8 8 8	2 6 2 6	[2] (3)	

* METASTABLE

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(1) - Saturation effect for 
$$\psi_{\text{th}} \equiv T \sim 4.10_{15}^{14} = 2$$
  
(2) - Saturation effect for  $\psi_{\text{th}} \equiv T \sim 2.10_{15}^{15} = 2$   
(3) - Saturation effect for  $\psi_{\text{th}} \equiv T \sim 10_{10}^{20} = 2$ 

TABLE II - GROUP 6

	Half-	Maximum	Or	der of a	agnitude			I	mpo	rt	anc	8		¥R	EN	DA re	ខប្	uest	5			Εv	7a]	luat	ed	
Isctope	life	yield	% (2200)	RIc	5 (30keV)	FSA	Th re	orm	al or	F	ast act	or	A11	There	a	1/E		Fast		Rez.	p.		d s	ita		
		<b>75</b>	barns	barns	mb.	щò,	J	W	B	J	U	C		\$	P	75	5	2	ρ	8	0	٤	c	8	L	
7 9 SE	STABLE	0.186(V3)	40.	30.	aga ::.): +:::+:::, +,::-+:::	20,	-					-		**************************************	Ť								c			2
\$72R	16. E H	5.9 (0 5)	0.2	1.		4.																	C			1
1120	STABLE	.197(PL9)	(0.5)	15.	250.	40.		}			}												C	8	1	4
11300	STABLE	.127(PU9)	20000.	400 <b>.</b>	700.	80 .	J	H	25				E 3	5	3		į					ε	C	8	L	(2)
1245N	STABLE	.119(PU9)	0.15	10.	25.	7.					}												C	8		ł
12558	2.7 Y	.192(PU9)	1.5	20.	300.	15.								3000	3								C			1
127TE #	169. 0	0.19(U 8)	3.	50.		25.								90Cb	3	]	1						C			i
133 I	20.8 H	6.8 (PU9)	<0.1	<0.1		<0.1								900b	2 ]9	x000 b/2							C	, ,		i
1368A	STABLE	0.15(PU9)	0.4	15.	80 .	15.												10	2				C	5		(1)
140LA	40.2 H	6.3 (0 5)	3.	60.		60 .										ł		i					C			1
156GD	STABLE	0.16(989)	(7.)	100.	500.	80.			i					5	1	5 h	. ]			10	2		C	8	L	Į
157GD	STABLE	0.11(PU9)	260000	1000.	1000.	100.		M			[		E	5	1	5 a	.]			10	2	E	(C)	6	L	(3)
man commune 1		whether the state of the state				The sum amounts when many		/	المسمعا	1	1 I	1 5	· (		- L -				_ 4			1	4 _ J	1 1	<b>_</b>	h

## * METASTABLE

(1) - Request both by reactor physicists as by astrophysicists.

(2) - Saturation effect for 
$$\varphi_{\rm spr} T \cong 5.10^{19} \, \rm cm^{-2}$$

(3) - Saturation effect for  $\psi_{\text{th}} \approx 4.10^{18} \text{ cm}^{-2}$ 

	Half-	Marinus	02	der of m	agnitude		Ī		Isp	201	rtar	100		¥)	RB	NDA re	equ.	st	3			B٧	val	ua	ted	
Isotope	life	yield	^G c(2200)	RIc	C (30koV)	FSA	Thre	ers	al or	re	ast	or	A11	Therm	al	1/5	F	at	Ι	Rea.	. P	•	da	ta		
		%	barns	baras	нb.	pb.	J	M	8	J	U	С		3	ρ	2	1		P	×.	P	٤	С	8	L	
785E	STABLE	0.098(U3)	0.4	6.	200 .	30.																	c	-		
9CZR	STABLE	6.5 10 31	0.1	0.2	15.	7 0					1						1						C	ø		
915A	9.7 H	5.7 (0 5)	0.15	0.6		2 .		Ì									1						C			
93 Y	10.2 H	6.8 (U 3)	0.1	1.9	!	2.								1		Ì							C			
111AG	7.5 D	0.371909)	3.	100 -		30.															11	1	C			
11400	STABLE	.094(PU9)	0.3	20 .	300.	50.				İ							{		1				C	8	L	
1158N	5.E4 Y	-085(PU9)	200.	3400.	1000 .	140 -			ł		1								1				C	82	Ł	
116CD	STABLE	.CO1(PU9)	Į.,	2 a	100.	10.				ļ													C			
117SN	STABLE	.084(PU9)	2.5	15.	400.	50.									{								C	8		
118SN	STABLE	.083(PU9)	(0.4)	7.	60 .	20.													i				C	8		
1195N	STABLE	.C66(PU9)	23	4 .	300 .	30.																	C	8		
1205A	STABLE	.064(PU9)	0.15	1.5	40 .	14 .				{	1			1				ĺ					C	8		
12158	STABLE	.066 (919)	6.	200.	700.	65.	1				İ							Í				l	C	8		
122SN	STABLE	.071	0.2	8.0	35.	14 .				1	1												C	8		
12358	STABLE	•086(PU9)	4.	130.	400.	40 .					1												C	8		
126TE	STABLE	0.30(PU9)	lo lo	11.	80.	13.								1			1						C	82	L	(1)
12758	3.8 D	0.59(0 3)	1.	15,		10.				l				40000	3								C			
135 I	6.7 H	6.4 (0 5)	(1.)	(2.)		0.3	J	M		1	1		E 3									E	C			
135XE	5.2 H	7.4 (209)	3.8+6	7000.		2.	J	1	1					5	2							E	C			
1378A	STABLE	6.6 (209)	5.	5.	100.	15.											1					- {	C	8		
156EU	13.2 D	0.15(PU9)	(800.)	1500.		400.		Ħ	l		[		E'3	700	3	1						E	C			
15660	STABLE	0.07(PU9)	3.	80 .	400.	50.								5	1	5 1				10	2		С	B	Ł	

TABLE II - GROUP 7

(1) -  $y_{eff} = 0.050 \ \% \ for \ ^{233} g$ 

	TA 8	LE	II	-	GROUP	8
--	------	----	----	---	-------	---

Teotope	Nolf-	Mayimua	Or	der of ma	gnitude	89 49 48 49 44 49 44		Iı	e po	rt.	anc	6		WR	EN	DA re	qui	est	8	• ••• ••• •		Ev.	alı	at	ed	
780.0094	life	yield	J. (2200)	RIc	C(30keV)	PSA	Th re	ac'	aal tor	r	Pas	t 201	A11	Thors	18 1	1/E		Pas	t	Res.	р.		iai	ta		
		\$	barns	barns	æb.	æb.	J	W	2	J	U	C		8	ρ	3	p	2	ρ	2	P	E	c	8	L	
87KR	760 M	3.1	(300.)	(130.)			J	W	[	1																(4)
96M0	STABLE	0.006(13)	1.2	25.	100.	20.			1		1			ļ	ļ			10	2	10	2		C	8		(1)
100RU	STABLE	-002(PU9)	6.	20.	200.	50.	3							10	2	25	2	1			}		C	8		
10220	STABLE	1.6-7	5.	2.	300.	70。	5		1	1	1		1					1						8		
1048H#	· ** 4 版	1.E-5	800.	(300.)						1	1						i									1
1048M	42. 5	1.8-5	40.	(300.)			J			1					ì				ł							1
105RU	4.4 H	5.1 (PU9)	0.2	5.		40 e		H H	}		1		183	1								E	C			
10746	STABLE	3.6 (PU9)	35.	100 °	800.	120.			1	!	1	1	183		i			l	i			£		3	1	
1090	13.5 H	1-95(b?a)	5.	60.		20.			1	}			1		Î	۲ ۲				10	F	E	C	_		(4)
11000	STABLE	1.8-5	(3.)	40 a	400.	80.					Į	ł	Į				.	10	2			1	C	8		(1)
13405	5°1 A	.0003(19)	140.	(200.)		100.	J		1		Ì	Í	j	10	5	25	2	i	ļ			_ 1	C		ł	{2}
1489 M	5.4 0	3E-419091	2500.	40000 -		250 .	ا قد	M	115				E3	5	1	10 p		l				Ξļ	C	ĺ	1	(2,3)
168940	41.5 0	36-4(909)	25000.	30000.		150.	3	11		i l	1	1	183					1	i			E	¢į	ļ		131
1485M	3.5141	6E-4(PU9)	5.	40.	250.	60.	1	뇛	1.				E3	_						10	2	E	Ň,	8	5	
15054	STABLE	.03519091	100.	260 a	400.	70 .	3	M	14	1			E 3	5	1	3				10	2	3	C	8		(2)
151PM	28.4 H	0.93(0 8)	200.	1200 .		150.		¥	1	1			E3	10	2	10	-			_		E	C	_		
15160	STABLE	0.9310 81	9000.	3200.	2500.	300.	] ]		1	1		1	1	5	2	10 2	2		İ	10	2	8		8		(4)
153SH	47. H	0.48(909)	(1000.)	3000.		(500.)		W	110				E3	4000	2	505	2	1				5	C			
154EU	706 Y	.00441 909	1500.	1500 .	3000.	700.		19	23		1	1	183	10	2	10	- 1				_	Ξi	c	. 4	L	(4)
155GD	STABLE	0.26(209)	60000-	1500.	2000 .	250 .					27		183			5 1				10	2	E	C	8	2	
15760	15. H	C.11(PU9)	(500.)	1300.	i i	200 -		Ħ		İ.			E3									ε	c			(4)

* NE TA STABLE

(1) - Requested both by reactor physicists as by astrophysicists.

(2) - Great production in thermal neutron capture.

(3) - Saturation effect in thermal neutron for  $\psi_{\text{th}} \approx 5.10^{14} (^{148}\text{Pm})$  and  $\psi_{\text{th}} = 10^{11} \text{s.cm}^{-2} (^{148\text{m}}\text{Pm})$ .

(4) - y_{eff} ≼0.005 \$

# 2.1. Examination of accuracies required on capture crosssections.

The absolute precisions requested should be the same (to within a factor 2) inside each groups of our classification, but this is not the case.

In the thermal range for example the demand in group 1 is 5% for  99 Tc ( $\tau = 2,1 \times 10^5$ a) and 10% for  143 Nd (st), which corresponds to precisions of  $\sim 1$  and  $\sim 70$  barns respectively; in group 2 the same applicant requires 10% for  107 Pd and  131 Xe, i.e. absolute accuracies of  $\sim 1$  and  $\sim 10$  barns. Three requests exist for  147 Nd : the accuracy desired for the first is 350 b whereas the 2nd and 3rd ask for 20% and 5% respectively, i.e. about 10 b and 3 b.

The same discrepancies appear for the capture resonance integral : in group 1 the relative precisions of 10% for  97 Mo and 20% for  99 Tc correspond to absolute precisions of  $\sim 2$  and  $\sim 40$  barns. In group 4 the accuracy requested for  132 Te ( $\tau = 3.3$  d) lies between 500 and 3000 barns according to the RI_ value, but is  $\sim 10$  b for  137 Xe ( $\tau = 5.3$  d).

Such scattering is essentially due to the fact that the requests are expressed in relative values, and always lie between 5 and 20%. This is especially true in WRENDA 73 for fast neutron  $\sigma_c$  and for the resonance parameters : the precision asked for is always 10% (except in one case :  $^{133}Cs$ ).

One WRENDA 73 applicant (WALKER) has expressed absolute precisions ; a group of three others (BAYARD, EHRLICH and SNYDER) requested relative precisions dependent on the absolute value. Each of these two groups of requests has an internal consistency but they differ greatly between each other about the demanded accuracy : the former asks for 600 b and 350 b respectively for  99 MO ( $\tau = 67$  h) and  147 Nd ( $\tau = 11.1$  d), while the latter require from 5 to 50 b for these two nuclei according to the absolute value of  $\sigma_{e}$ .

To avoid such inconsistencies and to give the re-
quests some meaning the accuracies on  $\sigma_r$  (2200) and RI_c should be expressed in absolute values ; it is also desirable that requestors harmonize their views on the needed accuracy.

### 2.2. Definition of a required precision.

The requests were not examined for purposes of criticism but in order to define the accuracies required. Despite the scatter of the values, averages given in table I were adopted. These values will be used in the following part of this review, on the understanding that they are averages for each group and that the individual precisions can vary as a result of differences in yield or half-life.

In addition the accuracies required are reduced when disappearance by capture is greater than that due to radioactive decay.

## 2.3. Effect of disappearance by capture.

When : 
$$\int \sigma_e \Psi dE = \sigma_e \phi > > \lambda$$
 and  $\sigma_e \phi T > > 1$ 

the nucleus disappears mainly by capture ; the precision on  $\sigma_e$ may then be poor without this affecting the permanent running of a reactor. Several cases of this sort exist, the best known probably being ¹³⁵Xe ( $\tau = 9.2$  h) for which the precision demanded in WRENDA 73 on  $\sigma_e$  (2200) is only 13000 barns. In the case of nuclei for which this saturation effect can arise table II gives :

- the value of  $\phi T = 1 / \overline{\sigma_c}$  if  $\tau > 2$  months, - the value of  $\phi = \lambda / \overline{\sigma_c}$  if  $\tau < \text{months}$ ,

 $\tau$  being the half-life and T the irradiation time.

## 2.4. Comments on the demands expressed in WRENDA 73

Several requests concern stable nuclei which have little importance as F.P.'s ; this applies for instance to 5% demand on  158 Gd (group 7), the demand on  110 Cd (group 8) etc...

These nuclei were probably not regarded as F.P.'s by

the applicants and the requests have been ignored, though some are aimed at improving the systematics.

### 3. EXISTING DATA.

### 3.1. Evaluated data.

The status of existing evaluated data is exposed elsewhere [Va 73], but a brief mention can be made of three data libraries recorded on magnetic tape :

- two are devoted entirely to F.P.'s : the Italian, which we shall call "BENZI" [Be69, Be71] , and the Australian which will be referred to as "COOK" [Co70, Ro71]. The former only gives capture cross-sections above 1 keV, together with  $\sigma_{\rm in}$  per level for a few nuclei ; the second is exhaustive (all reactions and all energies) but only gives the total inelastic cross-section.

- the third, E N D F / B-3, contains about fifty F.P's [Li 71].

Given the importance of these libraries their contents are listed in table II. Many other less well-known evaluations exist ; we can notice in the thermal field (including the resonance integral) :

- that of WALKER [Wa 72] who recommends values of q (2200) and/or RI for about 140 F.P.'s ;
- that of SAKATA and NAGAYAMA [Sa 73] who give values of  $\sigma_e$  and/or RI for more than 200 nuclei.

Other recent compilations exist but are seldom independent of the values recommended by WALKER ; this applies particularly to those of CLAYTON [C1 72] which are actually a revision of COOK'S data library, apparently not in circulation.

The contribution of POPE and STORY [Po 73] to this Panel is a compilation of values for :

- σ_c (2200)

- RI
- FSA, "fission spectrum average" for six fission product libraries recorded on magnetic tape, i.e :

- 2 versions of the Australian library : 1967 and 1971, we only use second version ;
- 2 complementary editions of the Italian library, which we consider together ;
- the E N D F / B-3 library ;
- the U K N D L library.

This last contains five evaluations independent of the other libraries (see table III).

The contribution of POPE and STORY also contains calculations or experimental values of  $\sigma_{c}$  (2200) and RI_c which we included in table III when they were not already present. Table III compares date on  $\sigma_{c}$  according to different authors.

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Table III: EVALUATIONS, GROUFS 1 - 8

#### Contents of Table III:

Capture cross sections at 0.0253 eV and at 30 keV taken from different nuclear data libraries.

Resonance integrals RI_C and fission spectrum average FSA cross sections calculated with these data.

Significance of the symbols used.

B : BENZI V. et al. [Be 69, Be 71]

C : COOK J.L., The Australian fission product library [Co 70]

E3 : Third version of ENDF/B

E31: RIc given in the ENDF/B3 comments

- E32: RIc calculated with ENDF/B3 data by POPE and STORY [Po 73]
  - J : From the Japanese Working Group.  $\sigma_c$  (0.0253 eV) and RI_c from SAKATA and NAGAYAPA [Sa 73],  $\sigma_c$  at 30 keV from MATSUNOBU [Ma 73]
  - L : LAUTENBACH [La 73], group cross sections library of 26 energy groups.  $G_c$  at 30 keV is a mean value between 21 and 46 keV.
  - S : SCHNITTROTH F. and SCHENTER R.E. [Sc 73a]
  - W: WALKER [Wa 73] The quoted values are the reduced resonance integral exepted for 105 Rh.

Isotope	The	rmal capture cr (bar	oss sect ns)	tion	R	sonan	ce inte	gral (	barns)	)	Cross	secti	on at	30 keV	(mb.)	Fi	ssion averag	spec ge (m	trum b.)	
	H	J	С	E3	W	J	c	E 31	£ 32	L .	c	8	s	J	ι	с	8	E3	L	
885R	0.005	0.005+- 0.001	.0054		0.06	0.057	0.047	T		0.067	3.0	26.			3.05	2.8	14.5		2.0	+
9 C SR	0.80	1.00+- 0.60	0.795			2.00	0.40			0.52	26.			17.0	26.5	4.0	140/		3.0	ļ
92ZR	0.26	0.25+- 0.12	0.25		0.50	0.58	0.29			0.64	5.0	36-	40.		5.6	4.5	0.2		6.0	
93Z.R	2.00	1.50	1.99		22.0	1.80	25.9			8.00	226.	144.	90.	203.	231	7.6	11.0	1	20.0	
94ZR	0.070	C. 075+- 0.008	0.076		0.26	0.28	0.21			0.35	17.0	30.	20.		17.2	2.9	7.6		7.0	1111
96ZR	0.20	0.050+- 0.010	0.196		6.00	5.44	5.30			5-90	10.0	28-	44		9.8	5.8	8.0	1	14.0	121
97M0	2.20	2.20+- 0.70	2.20	1.90	15.0	14.5	15.0	15.8	16.1	15.0	475.	378.	371	345.	485	30.0	41.8	39.0	43.0	101
9 8MO	0.14	C.150+- 0.200	0.145	0.15	8.00	6.73	6.68		6.80	7.00	166	104.	94		169	37.0	27.3	29.0	16.0	1233
99TC	22.0	22.0+- 3,00	22.0	19.0	200.	204.	197.	1	3 5 3.	368.	981	844		950.	1000	46.0	93.0	58.0	105.	1
100MO	0.20	0.50+- 0.50	0.225	0.20	3.90	3.94	6.53		3.62	8.40	149.	71 -	74-		152	15.0	14.7	16.0	19.0	1151
101RU	5.20	3.30+- 0.90	4.84	5.20	76.0	73.8	85.7	84.9	79.6	82.0	1094.	901	850.	1382.	1116.	287.	74.5	73.0	125.	16)
102R U	1.30	1.44+- 0.16	1.29	1.30	4.20	10.6	10.6	4.08	6.90	14.5	378.	348.	244.	333.	385.	58.0	86.5	95 -0	39.0	1.01
104RU	0.47	0.47	0.44	0,47	4.40	4.60	5.41	5.43	3.73	10.1	387.	155.	160.	165.	395	16.0	24.6	21.0	39.0	
10590		11.0	11.0	14.0	85.0	80.5	74.5	86.3	86.3	90.8	1217.	1 221 .	984	1460.	1241 .	57.0	106.	120.	146.	
132XE	0.49	0.272	0.461		2.60	0.122	2.44			1.60	74.	116.	60.		76 .	28.0	27.0	1	9.0	
13 3C S	29.5	29.0+- 1.5	29.5	29.5	450 .	440.	377.		3 80.	387.	686-	693		660.	700	38.0	55.0	73.0	72.0	143
134XE	0.263	0.228	0.253		6.10	0.302	0.260			5.40	47.	60.5	9.0		477	6.1	17.0		5.0	175
135C S	8.90	8,70+- 0.50	8.79	8.90	58.0	80.5	58.1	58.3	30.2	20.5	248.	294 .	64.	345-	253.	110.0	27.0	6.0	28.0	1
136XE	0.200	0.281+- 0.028	0.199			0.40	0.115	1		0.16	9.0	9.3	3.0		9.2	2.2	4.5		1.0	1
137C S	0.110	0.110+- 0.033	0.103	0.11		0.08	0.411		0.23	0.60	23.	36.	8.0	25 .	23.	2.2	4.3	1.0	1.0	
1388A	0.40	0.35+- 0.15	0.35		0.20	0.163	0.210			0.17	2.0	6.0			2.14	3.9	27.4		1.0	l
139LA	9.00	8.20+- 0.80	9.00	9.20	11.0	13.3	15.3	12.4	16.2	18.2	56.	40.	48.	1	57	5.8	6.9	7.0	5.0	
140CE	0.59	0.60+- 0.06	0.59		0.24	0.49	0.489	1		0.35	20.	25.	22 -		20	10.7	17.3	1	2.0	1
142CE	0. 90	0.95+- 0.05	0.858		1.00	1.00	1.49	1		1.00	49.	47.	43.	ł	50.	8.6	14.2		6.0	1
143ND	325.	335.+- 10.0	325.	318.	60.0	202.	64.5	87.0	136.	137.	185.	339.		360.	189.	37.0	42.3	37.0	28.0	

TABLE III GROUP 1

1) -  $\sigma_{th} = 0.06 \pm 0.01$ ,  $RI_c = 0.37 \pm 0.04$  [Ed 73] 2) -  $\sigma_{th} = 0.006 \pm 0.001$  [Ed 73] 3) -  $\sigma_{th} = 0.13 \pm 0.01$ ,  $RI_c = 7 \pm 1$  [Ed 73], g = 1.024) -  $\sigma_{th} = 29.5 \pm 2.0$ ,  $RI_c = 450 \pm 20$  [Ed 73]  $RI_c = 460$  [Cl 72] 5) -  $RI_c = 4.0$  [Cl 72], g = 1.016) -  $\sigma_c(2200) = 5.17$ ,  $RI_c = 79.6$  [Cl 72] 7) -  $RI_c = 4.6$  [Cl 72] 8) - g = 1.01

TABLE III GROUP 2

	The	rmal capture ci (bai	oss sectors)	tion	Re	sonand	e inte	gral (	barns)		Cross	section	n at 3	0 keV	(mb.)	Fisa	ion s erage	pectr (mb.	-um )	
Isotope	W	J	c	E 3	W	J	c	E 31	E 32	L	с	я	S	J	L	с	8	E3	L	
86KR 87RB 89SR 89SR 91Y 91ZR 95ZR 95ZR 95ZR 103RU 103RU 103RH 106RU 107PD 108PD	1.00 0.120 0.420 1.28 1.40 1.60 14.5 150. 0.150	$\begin{array}{c} 0.060+-0.020\\ 0.120+-0.030\\ 0.500+-0.100\\ 1.28+-0.02\\ 1.07\\ 1.58+-0.12\\ 14.5+-0.5\\ 150.+-5\\ 0.146+-0.045\\ 10.0\\ 12.3\\ \end{array}$	0.998 0.120 0.416 1.28 1.05 1.58 0.488 14.5 7.70 149. 0.137 9.41 12.0	5.00 14.3 5.00 148.2 0.125 5.00	2.30 0.30 6.50 100. 1100. 2.00 240.	0.07 2.30 0.80 0.765 1.40 7.70 106. 1070. 1.30 36.0 224.	0.460 2.47 0.35 0.66 1.92 7.78 5.40 106. 106. 1.28 80.1 215.	7.43 107. 2.25 1048. 1.27 78.2	7.98 109. 9.24 1048. 1.41 79.7	0.08 2.10 0.78 8.10 117. 1038. 8.10 68.0 236.	16.0 35. 34. 124. 414. 804. 356. 1198. 424.	8.7 27. 23.4 80. 336. 793. 1245. 188.	29. 44. 178. 392. 844. 77. 894. 185.	355. 1175. 120. 1440.	16.6 35.5 34.6 126. 422. 820. 363. 1222. 432.	1.6 3.2 2.2 2.6 6.2 8.6 5.6 39. 59. 82. 12.0 76. 13.0	2.5 3.25 5.6 14.2 36.5 85. 121. 32.1	13.0 36. 42. 92. 12.0 88.	2.0 3.0 3.0 10.0 36. 93. 37. 123. 38.	(3) (2) (4) (7) (5)
131XE 141CE 141PR 144CE 145ND 146ND	100. 29.0 11.0 1.0 45.0 1.40	110.+- 20. $29.0+- 3.0$ $12.0+- 3.0$ $1.00+- 0.10$ $52.0+- 2.0$ $10.0+- 1.0$	110. 28.8 11.0 0.930 44.9 1.31	29.0 11.4 41.2	830. 13.0 2.20 250. 2.00	784. 13.0 18.0 2.62 306. 25.0	28.7 17.9 2.58 272. 2.32	29.1 18.4 293.	29.1 18.8 298.	898. 19.1 237. 2.90	358. 176. 298. 85.	491. 139. 418. 138.	35.	85. 440.	180 • 304 • 87 • 1	45. 13.0 15.5 7.6 25. 23.	39. 19.0 32.4 30.1	13.0 16.0 25.	23. 46. 14.0	(6) (1)

(1) ~ 
$$\operatorname{RI}_{c}/\operatorname{G}_{c(2200)}^{*} = 1.42 \pm 0.10$$
 (Ri 73).  
(2) -  $\operatorname{RI}_{c}(\operatorname{COOK}) = 116$  (Po 73)  
(3) -  $\operatorname{G}_{c}(\operatorname{Pile})^{*} = 1$  (Ed 73)  
(4) -  $\operatorname{G}_{c}(\operatorname{Pile})^{*} = 10$  (Ed 73)  
(5) -  $\operatorname{G}_{c}(\operatorname{Pile})^{*} = 0.15 \pm 0.05$ ,  $\operatorname{RI}_{c} = 2.0 \pm 0.6$  (Ed 73)  
(6) -  $\operatorname{G}_{c}(\operatorname{Pile})^{*} = 29 \pm 3$  (Ed 73)

(7) - g = 1.02

TABLE III GROUP 3

Tsotone	The	ermal capt	ure cı (baı	oss sec ns)	tion	Re	sonanc	e inte	gral (	barns)		Cross	sectio	n at 3	50 keV	(mb.)	Fis	sion verag	apect: e (mb	rum .)	
	W	J		C	E3	ω.	J	c	E 31	E 32	L	C	B	S	J	L	с	B	E3	L	
83KR 84KR 85RB 95NB 109AG 128TE 129I 130TE 14CBA 143PR 144ND 147PM 148ND 149SM	200. C.100 0.42 4.0 92.0 0.215 28.0 0.22 1.60 100. 3.60 170. 2.50 42000.	205.+-0.1600.764.091.0+-0.17028.0+-0.261.57+-89.0+-5.00+-200.+-2.90+-41000.+-	30. 3.0 3.0 0.03 10.0 0.60 50. 0.50 2000.	200. 0.097 0.42 1.45 95.0 0.213 28.0 0.26 1.58 97.7 3.39 170. 2.35 42040.	187. 4.0 92.0 100. 170. 41190.	150. 8.0 7.0 1450. 1.58 23.0 0.40 13.0 150. 3.6 2200. 20.0	245. 3.54 3.2 1460. 10.0 36.0 3.0 13.6 190. 817. 2210. 18.7 3260.	211. 3.59 25.0 1442. 1.55 25.5 0.178 13.6 472. 7.57 2156. 14.0 3594.	231. 26.1 157. 2199.	2 35. 26.6 1457. 167. 22 36. 31 83.	232. 3.70 4.3 1470. 1.98 43.0 0.39 5.10 2270. 2.60 5541.	418. 27. 282. 1470. 55. 445. 4.0 72. 897. 114. 1248.	642. 30. 289. 897. 29. 13.0 92. 999. 168. 1697.	143. 286. 443. 459.	1530. 640. 112. 755. 1400.	426. 27.3 287. 1499. 55.7 454. 3.89 73.2 915. 116. 1273.	39. 6.1 20.7 14.0 91. 3.5 47. 2.3 14.0 38. 33. 124. 20. 123.	61.8 7.05 26.7 130. 6.9 50. 3.2 27.1 83. 29.9 168.	37. 28. 64. 42. 138.	41. 3.0 27. 174. 6.0 60. 3.0 11.0 155. 20. 182.	<pre>(1) (9) (2) (6) (7) (3) (3) (4) (8)</pre>
(1) - G	(2200) ⁼	206, RI c *	253, F	1.13 SA = 61?	(Po 73)	UKNDL	- 0- c(t	$(2.56)^{-200}$	± 10, 1	RI _c = 150	$\frac{16.3}{1}$	192. Ed 73)	361.			1958.	25.	53.3		35.	(5)
(2) - 0	 (2200) ⁼	93.6, RI c	= 1454,	FSA = 62	2.5 (Po 7	3) UKND	L - <del>C</del> (	th) ^{= 93}	± 5, R	L _c = 150	00 ± 200	(Ed 73),	,g = 1.0	1							
(3) - 0	_ (2200) ⁼	188, RI _c =	2237, F	SA = 100	(Po 73)	UKNDL -	c(th)	= 182 ±	20, RI	= 2400	± 300 (	Ed 73),g	= 0.99								
(4) - F	$RI_c/\sigma_c(2)$	200) = 4.22	± 0.14	(Ri 73)			- •	•.													
(5) - H	RI _c / <i>d</i> _c(2)	200) =13.7	± 0.8 (	Ri 73)	•				••												
(6) - J	- (th) ^{=1.5}	57 [±] 0.03, 1	RI _c = 1	3 ± 2 (Ed	1 73)																
(7) - 5	- (th) ^{= 1(}	00 ± 10, RI	c ≖ 150	± 30 (Ed	1 73) - R	$I_c = 19$	0 (C1 7	2), g = (	0.99												
(8) - J		42100 ± 400	0 (Ed 7	3), g = 1.	.64																
(9) - RJ	د _د ≖ 7.0	(C1 72).																			

TABLE III GROUP 4

	The	ermal capture cr (bar	ross section rns)	R	sonaac	e inte	gral (	barns)		Cross	sectio	n at 3	iO kev	(mb.)	Fis a	sion verag	spect e (mb	ru <b>n</b> .)	
TROLODE	W	L	C E3	н	J	C	E31	E 32	L	C	8	s	L	Ł	c	8	E3	ι	ĺ
825E	0.045	2.10+- 1.50	1.73 5.	0.090	0.980	24.8		25.9			68.				21.	12.0	22.		
10690	0.300	0.292+- 0.029	0.285 0.2	0 5.60	5.96	8.30	11.9	10.1	10.7	339 <b>.</b> 280.	240.	187.		346 .	23.	46.9	41.	31.	(3)
1271	6.20	6.20+- 0.20	6.18	150	149.	152.	6.44	10.1	153.	653.	800.	690.		666.	73.	82.	24.	90.	
132TE	100	190.	0.0024		1700	C.007	57.4	40.9							0.2		<u> </u>		111
147ND	1744	1700	49.3 50	0	1100	644.	645.	648.	22.22	1 07 /					49.	167	51.		167
1515M 1525M	206.	210.+- 10.	206. 20	. 3100	2700.	1770.	3360.	3460.	3115.	600.	496-		2029.	612.	351. 56.	86.	401. 57.	81.	(2)
1) -	0- (Pi	$1_{0} = 190 \pm 90$	[Ed 73], RI	(COOK)	= 49,1	[Po 7	3]		'		40 au	inter als an old the	<u>.</u>	10 August 100 August 100 August 100	·	*** *** *** ***		*	

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1) 
$$-\sigma_{c}^{-}$$
 (Pile) = 190 ± 90 [Ed 73], RI_c (COOK) = 49.1 [Po  
2)  $-\sigma_{c}^{-}(2200)$  = 15000 ± 1800 [Ed 73], g = 0.90  
3)  $-RI_{c}$  = 5.6 [Cl 72], g = 1.01  
4)  $-\sigma_{c}^{-}(2200)$  = 0.28, RI_c = 7.1 [Cl 72]  
5)  $-RI_{c}$  = 2180 [Cl 72]  $\sigma_{c}^{-}(2200)$  = 200 à 300 b [Ma 73a]  
6)  $-\sigma_{c}^{-}$  (Pile) = 100 [Ed 73]

TABLE III GROUP 5

Tentone	The	rmal capty	ire cro (barn	ss sect s)	ion	Re	sonanc	e inte	gral (	barns)		Cross	sectio	n at 3	0 keV	(mb.)	Fis	sion verag	spect: e (mb	rum .)	
1901056	W	J		c	E3	W	J	С	E 31	E 32	L	С	B	S	L	L	с	ß	E3	L	
805E 818R 85KR 105RH 111C0 126SN 129TE 143CE 147SM 149PM 153EU 154SM 155EU	0.580 2.40 8.00 17000. 23.0 6.00 55.0 1400. 450. 5.00 4200.	0.610+- 3.10+- 8.00 18000. 24.3+- 0.300 6.00+- 87.0+- 1700.+- 390.+- 5.50 14000.+-	0.060 0.40 0.3 0.70 60.0 300. 80.	0.574 3.00 7.93 17000. 23.0 0.297 0.37 5.60 54.8 1390. 449. 4.70 14000.	17000. 5.00 57.0 1450. 453. 4040.	1.30 50.0 17000 47.0 600. 1500.	0.760 54.8 81.0 16500 51.0 2.70 624. 765. 1203. 16.0 7000.	0.986 59.5 8.02 16960 45.5 7.40 566. 917. 1203. 38.1 1223.	18580 616. 832. 1980.	623. 807. 1512. 1817.	60.6 9.40 69.0 601. 1635. 27.0 3629.	434. 178. 841. 670. 2460. 321. 1770.	32. 332. 610. 1236. 2702. 448.	582.	1080. 2400. 2200.	443. 181. 857. 683. 2514. 327. 1809.	7.3 39. 11.0 21. 23. 3.7 11.0 9.8 133. 92. 234. 47.8 153.	5.43 26.3 54. 209. 303. 69.7	63. 134. 68. 160. 620.	53. 16.0 88. 101. 423. 43. 283.	(2) (3) (4) (7) (5) (1) (6)

 $(1) - \sigma_{c(2200)} = 444$ , RI_c = 1500, FSA = 152 (Po 73) UKNDL,  $\sigma_{c(2200)} = 450 + 20$  (Ed 73), RI_c = 1279 (Cl 72), g = 0.98

(2) 
$$-\sigma_{-(\pm\pm)} = 2.7 \pm 0.2$$
, RI = 55  $\pm 5$  (Ed 73)

$$(2) - \sigma_{c(th)} = 2.7 \div 0.2, RI_{c} = 55 \div 5 (Ed 73)$$

$$(3) - \sigma_{c(th)} = 17000 \stackrel{+}{-} 2000, RI_{c} = 17000 \stackrel{+}{-} 3000 (Ed 73), RI_{c} (COOK) = 9880 (Po 73), g = 1$$

$$(4) - RI_{c} = 0.12 (Ed 73)$$

- (4)  $RI_c \approx 0.12$  (Ed 73)
- (5)  $\sigma_{c}(Pile) = 1400 300$  (Ed 73)
- (6)  $\overline{\sigma_c(\text{Pile})}$  = 4040 + 125 (Ed 73), g = 1
- (7)  $-\sigma_{c(th)} = 61 \stackrel{+}{-} 7$ , RI_c = 646  $\stackrel{+}{-} 60$  (Ed 73)

TABLE III GROUP 6

	The	rmal capture cr (bar	oss sec ns)	tion	Re	sonan	ce inte	gral (	barns)		Cross	sectio	n at 3	0 keV	(mb.)	Pie	saion ivera	spect se (mi	rum ().)	
180.000	W	L. L.	с	E3	W	Ŀ	c	E 31	E 32	L	с	В	s	J	L	С	8	E3	L	
795E 972R 112CD 113CD 124SN 1255B 127TE 133I 136BA 140LA 156GD 157GD	2.00 19940. 0.134 0.400 2.70 1.40 264000	40.0 $0.030+-0.015$ $2000.+-300.$ $0.140$ $1.56$ $0.400+-0.400$ $3.10+-1.00$ $11.5+-7.2$ $254000+-2000.$	39.3 0.202 1.99 20190. 0.165 0.97 2.75 0.003 0.395 2.51 6.14 264000	19900.	17.0 9.03 17.0 70.0 90.0	18.0 11.8 530. 11.0 23.0 10.2 60.0 105. 515.	54.8 1.54 14.1 365. 11.3 19.0 48.2 0.005 17.1 70.6 121. 3410.	835.	3 E 6 . 11 46 .	16.4 490. 107. 1108.	290. 866. 369. 905.	245. 631. 26. 80. 634. 1392	329.		2 <b>96.</b> 883. 376. 923.	19.0 4.0 35. 33. 5.6 15.0 25. 0.05 14.0 60. 45. 56.	56.2 81.2 8.13 19.6 117. 101.	79 <b>.</b> 102.	28. 88. 69. 148.	(1) (2) (4) (3)
1) - 2) -	G(220)	$(2) = 26601, RI_{c}$ = 2.7 ± 0.3, R	= 309, I _c = 69	$FSA = \frac{1}{2}$ $\pm 4 \left[ E \right]$	58.2 [1 1 73]	°o 73]	UKNDL,	RIc	(COOK	) = 28	9 [Po 7]	3], g	1.33	**	*****					<b></b>

3) - 
$$RI_{c}(COOK) = 3160$$
 [Po 73],  $g = 0.85$ 

4) -  $RI_c = 90.0 [C1 72]$ 

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TAB	LE	111	GROUP	- 7
				-

Tentone	Therma	i capture	crose barns)	section	1		Resona	nce int	egral	(barn:	в)	Cross	sectio	n at 3	0 keV	(mb)	Fis ave	sion rage	spect (mb)	rum	
1300000	W	L		c	E3	W	J	с	E31	E 32	L	c	8	S	J	L	c	B	E3	L	
785E 90ZR 91SR	0.40 0.10	0.38+- 0.10+-	0.04 0.07	0.40 0.10 0.148 0.078		4.30 0.150	6.65 0.268	7.08 0.079 0.616					183. 16.0				13.5 5.0 2.40	37.2 9.2			
111AG 114CD 115IN	3.00 0.34 206.	0.30+- 199.	0.15	2.82 0.325 203.		100. 23.0 3340.	15.9	106. 16.4 3212.			20. 3287.	349. 1 023.	317. 761.			356 • 1044 •	35. 29. 101.	76.8		34. 123.	(2)
1175M 1185N 1195N	1.40 0.80 1.20	2.70 0.010+- 2.50	0.006	1.40 0.836 1.20		12.0 8.20 3.50	16.8 5.35 5.00	16.5 7.39 5.28					416. 65. 293.				48. 14.0 36.	46.8 24. 29.6			
1205N 1215B 1225N 1235B	0.20 6.20 0.18 4.20	0.14+- 6.55 0.18+- 4.10	0.03	0.141 6.19 0.18 4.18		200 • 0 •60 1 30 •	1.50 205. 0.92 124.	1.30 205. 0.884 126.					42. 721. 36. 390.	721.			12.0 63. 13.0 36.	15-8 66.9 13.6 42.			(3) (4)
126TE 127SB 1351	1.00	1.04	١٩.6	1.00 0.92 0.022	15.0	10.0	11.8	8.16 14.7 0.03	6.77	6.58	12.5	97.	74.			991.	11.0 9.40 0.26	16.0	0.	11.0	
1378A 1378A 156EU 158GD	5.10 2.8	5.10+- 3.5+-	0.40	5.10 481. 2.67	2000.	2.00	5.65 135.	4.83 1258. 97.8	1947.	1947.	62.9	240.	106. 525.			245.	2.1 15.0 319. 36.	14.4 87.3	567.	41.	(1)

(1) -  $RI_c = 5893$  (Po 73) UKNDL,  $RI_c = 7620$  (C1 72), g = 1.16

(2)  $-\sigma_{c(th)} = 3 \stackrel{+}{=} 2$ , RI_c = 105  $\stackrel{+}{=} 20$  (Ed 73) (3)  $-\sigma_{c(th)} = 6.2 \stackrel{+}{=} 0.1$ , RI_c = 200  $\stackrel{+}{=} 10$  (Ed 73)

- (4)  $-\sigma_{c(th)}^{-} = 4.2 + 0.2$ , RI_c = 120 + 10 (Ed 73).

	TA	61	-E	11	I	GROUP	8
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	The	rmal capt	ure cr (bar	oss sec ns)	tion	R	esonan	ce inte	gral (	barns)		Cross	sectio	n at 3	0 kre∛	(mb.)	Fi	ssion avera	spect ge (mt	rum .)	
lsotope	¥	J		ſ	£3	W	J	C	E31	E 32	L	C	В	S	J	L	c	8	E 3	٤	
87KR 96M0 100RU 102PD 104RH* 104RH	1.20 5.80	<600. 1.20+- 10.4+- 4.80+- 800.+- 40.0+- 0.20+-	0.60 0.7 1.50 100. 30.0	1.20 5.49	0.30	25.0 9.00	<270. 26.3 11.3 2.20 360. 360.	26.1 41.2	5-12	7 . 85			107. 181. 319.				18.0 81. 41.	25.2 34.1 75.2	40-		{7}
107AG 107AG 109PD 110CD	10.0	35.0+- 0.10+- 134.+-	5.0 0.03	5.25 9.98	36.8 5.11	37.0	103. 43.4 1150.	115. 60.8 50.9 86.1		60.2			828. 372.				29. 63. 99.	105. 102.	118. 15.0		(1)
148PM 148PH* 1485M 1505M	3000. 24000. 4.70 100.	2000.+- 30000.+- 9.00 102.+-	1000.	2995. 27000. 4.42 102.	2200. 24000. 4.73 100.	240.	48000 35000 50.0 227.	43950 32000 18.4 329.	44060 31000 53.9 240.	36440 10570 54.2 252.	23.0 422.	255. 403.	251. 410.			260. 411.	188. 189. 60. 60.	65.3 85.1	272. 124. 61. 61.	34. 76.	(3) (4) (6)
151PM 151EU 153SM 154EU	400-	8800.+-	100.	173. 335. 1380.	150. 9350. 9990. 1500.		3300 <b>.</b> 7900 <b>.</b>	1205. 1110. 1250.	1197. 5460. 1320.	1201. 3250. 5459. 1321.	1544.	2740.	2 507.				120. 158. 750.	360.	122. 218. 1685 787.	484.	(2) (5)
155GD 157EU	66 000.	61000.+-	1000.	61000. 191.	61100. 2000.		1560.	1560. 824.	1480. 1640.	1555.	1685.	1740.	2168.			1778.	102.	158.	332. 399.	271.	(8)

* Metastable.

1)  $-\sigma_{c}(2200) = 36.9$ , RI_c = 115, FSA = 114 [Po 73] UKNDL 2)  $-\sigma_{c}(2200) = 8336$ , RI_c = 2390, FSA = 207 [Po 73] UKNDL, g = 0.893)  $-\sigma_{c}$  (Pile) = 3000  $\pm$  2000 [Ed 73], RI_c (COOK) = 25700 [Po 73] 4)  $-\sigma_{c}$  (Pile) = 25000  $\pm$  2000 [Ed 73], RI_c (COOK) = 19300 [Po 73],  $\sigma_{c}(2200) = 24000$  [Cl 72] 5)  $-RI_{c}$  (COOK) = 1160 [Po 73], g = 1.076)  $-RI_{c} = 257$  [Cl 72] 7)  $-RI_{c} = 11.6$  [Cl 72] 8) -g = 0.84

#### 4. EXAMINATION OF CAPTURE CROSS-SECTION DATA .

The accuracies necessary for each group of nuclei are defined above. The quality of existing data will be examined against three criteria :

- 1) compliance with the precisions necessary ;
- compliance with the precisions necessary improved by a factor 2 ;
- absence of data or abnormal disagreements between recommended values.

There is no need to comment on the first criterion, the second being a projection into the future. The third is governed by the following two ideas :

- in the absence of experimental data the recommended value, if it exists, may be completely wrong ; this is especially true for  $\sigma_{2200}$ .
- large disagreements on the recommended values are inacceptable even if this does not immediately affect the calculation of the capture rate, given the errors which exist on other nuclei.

### 4.1. Thermal field.

The values used are chiefly those recommended by WALKER on the one hand and by SAKATA and NAGAYAMA on the other. These are two independent works ; unfortunately WALKER does not give the error on the recommended value, and SAKATA and NAGAMAYA do not give their sources of information.

As a general rule the precisions requested in WRENDA are adopted for group 8, although they sometimes seem exaggerated : 100 barns for  148  Pm ( $\tau$  = 5.4 d) for example; table IV shows the result of the data examination. We conclude that :

 the required precision is not obtained for 15 nuclei at most; on 9 of these its value is not well known because of the saturation phenomenon.

## TABLE IV

GROUP	Accuracy required(2)	Nucleus	Half life	Obtained accuracy	
I II	5 b 10 b	¹⁴³ Nd ¹³¹ Xe	st st	10 b 20 b	
111 IV	20 b 20 b 7 7	83 _{Kr} 147 _{Pm} 149 _{Sm} 151 _{Sm}	st 2,6 y 10 ¹⁵ y 93 y	30 b 50 b 1000 b 2000 b	
v	100 b ? 7	149 _{Pm} 155 _{Eu} 105 _{Rh}	53 h 5 y 35,5 h	300 Б 4000 Б 3000 Б	(1)
VI VII	? ? 500 b	157 _{Gd} 135 _{Xe} 156 _{Eu} 148 ₅₀	st 9,2 h 15,2 d	5000 b 70000 b 1000 b	
VIII	100 B (2) ? 4000 b	^{Pm} 148 _{Pm} = 153 _{Sm}	5,4 a 41,5 d 47 h	3000 b 9000 b	

Capture cross section requirements unsatisfied. Thermal range 1) Obtained error greater than the required accuracy

# Obtained error greater than the required accuracy improved by a factor 2.

I	2,5 b	99 _{TC}	2,10 ⁵	3 b
	2,5 b	105 _{Pd}	st	5 b
V	50 b	153 _{Eu}	st	80 b
VI	?	113 _{Cd}	st	300 b

(1) - According Walker, the discrepancy is solved and accuracy is 125 b [R.S. MOWATT, Can.J. Phys, 48,1933(1970)] - But only one measurement.

(2) According WALKER, "all the saturating large cross section nuclides are well enough known for neutron absoption calculations except for  $^{105}\rm Rh$  and  $^{148}\rm Pm^m$ ; some indication of how the cross section varies with neutron temperature (or resonance parameters) would be useful.

- 2) if the precision needs to be better by a factor 2, only
   4 extra nuclei join the above list of 15.
- 3) no data exist for a large number of F.P.'s (WALKER mentions nearly 50) : we have only taken fifteen, those with the highest equivalent vield, and 7 cases are pointed out where disagreement exceeds a factor 5.

# TABLE IV

Capture cross section requirements unsatisfied. Thermal range 1) Obtained error greater than the required accuracy

GROUP	Accuracy required	Nucleus	Half life	Obtained accuracy
I	5 b	143 _{Nd}	st	10 b
II	10 b	¹³¹ Xe	st	20 b
III	20 b 20 b	83 _{Kr} 147 _{Pm}	st 26 v	30 b 50 b
	?	149 _{Sm}	$10^{15}$ y	1000 b
IV	;	151 _{Sm}	93 y	2000 b
v	100 b	149 Pm	53 h	300 b
	?	155 _{Eu}	5 у	4000 b
	?	105 _{Rh}	35.5 h	3000 b
VI	?	157 _{Gd}	st	20000 b
VII	?	135 Xe	9.2 h	70000 ь
	500 b	156 _{Eu}	15.2 d	1000 b
VIII	100 b (?)	148 _{Pm}	5,4 d	1000 b
	?	148 m Pm	41.5 d	3000 b
	4000 b	153 _{Sm}	47 h	9000 b
1				

2) Obtained error greater than the required accuracy improved by a factor 2.

I	2.5 b 2.5 b	99 _{TC} 105 _{Pd}	2x10 ⁵ y st	3 b 5 b
V	50 b	153 _{Eu} 113 _{Cd}	st	80 b
VI	1	Cd	st	300 b

# TABLE IV (contid)

# 3) Large disagreements or absence os data.

Group	Accuracy required	Nucleus	Half life	Precision obtained	
II	10 b	86 _{Kr} 95 _{Zr} 103	st 63 d	no data ""	
III	20 р	и 146 _{Nd} 95 _{Nb}	40 d st 35 d	factor 7 disagreement only1 inaccurate datum	
IV	50 b	¹⁴⁰ Ba 82 _{Se} 99 _{Mo} 132 _{mo}	13 d st 67 h 78 b	factor 20 disagreement no data	
v	100 b	147 _{Nd} 111 _{Cd} 147 _{Sm}	11 d st 10" y	no data no data factor 12 disagreement large disagreement (Wa 73)	
VI	200 Б	92 Zr	17 h	no data	
VII	500 ъ	$112_{Cd} \\ 127_{Te} \\ 125_{Sb} \\ 133_{I} \\ 156_{Gd} \\ 91_{Sr} \\ 93_{V}$	st 109 d 2,7 y 21 h st 9,7 h	factor 20 disagreement no data no data no data factor 8 disagreement no data	(1
VIII	30 b (Wr) 500 b (Wr)	118 _{Sn} 127 _{Sb} 135 _I 156 _{Eu} 151 Pm 151 _{Eu}	10,2 n st 3,8 d 6,7 h 15,2 d 28,4 h st	no data factor 50 disagreement no data no data no data and disagreement factor 2 disagreement 500 b disagreement	

For details about the discrepancies the reader can refer to table III. (1) According WALKER, no discrepancy between experimental data.

#### 4.2. Resonance Integral.

A new difficulty arises here, that of the resonance integral (RI_c) calculation from differential data. Small numerical errors are to be expected, but larger ones often arise. This is illustrated by columns 9 and 10 of table III, which give the RI_c values for the E N D F / B 3 library according to two different sources. The agreement is generally satisfactory apart from a few abnormal cases :  102 Ru,  104 Ru and  143 Nd in group 1 for example,  103 Ru in group 2.

We shall base on the recommended data given in table III, not one of which bears an error. The Japanese give several values, but often without the reference. LAUTENBACH only takes into account the resonance parameters (resolved range) or calculations by a statistical model (unresolved or continuous range), using the experimental resonance integral (RI_c) values to correct the parameters of the statistical model only when no values exist for the parameters of the first resonances.

Table V gives the result of the data examination.

Not all the values have been retained ; for example the Japanese  $RI_c$  value for  $^{133}Xe$  (group 4) is left out, being based on a COOK result dating from 1966 which was dropped in the latest version of the Australian library.

We conclude that :

- no more than 9 isotopes fail to meet the precision requirements;
- this number is increased by 8 if the precision needs to be twice as good ;
- 3) the recommended data are more scattered than in the thermal field : for many nuclei the resonance integral is the result of calculations based on the statistical model and the mean parameter systematics.

Very often an evaluation would suffice to resolve the

#### TABLE V

Car	pture	cross	sectio	on requ	lireme	nts	unsatisfi	led -	Resonance	integral
1)	Error	obtai	ined gr	reater	than	the	required	accu	racy.	

GROUP	Accuracy required Nucleus		Half life		Accuracy obtained	Comments
I	25 b	99 _{Tc}	2,1.10	5 a	50 b	
	25	143 _{Nd}	st		40	
III	100 (?)	149 _{Sm}	$\sim 10^{15}$	а	300	saturation forth
IV	250 (?)	151 Sm	93	a	700	saturation forth
V	500 (?)	¹⁰⁵ Rh	36	h	1000	saturation forth
	500 (?)	155 _{Eu}	5	a	3000	saturation forth
VI	1000	¹² /Te	34	j		no data
	1000	133 _I	20,8	h		no data
	1000	⁹⁷ zr	16,8	h		no data

2) Error greater than the required accuracy improved by a factor 2

I	12 b	93 _{2r}	st	20 b	
		¹³³ Cs	st	25	
		135 _{Cs}	2,3.10 ⁶	30	
II	25	107 _{Pd}	7,10 ⁶	25	
		¹³¹ Xe	st	50	
		145 _{Nd}	st	30	
III	50	147 _{Pm}	2,6	100	
IV	120	¹⁵² Sm	st	200	

3) Large disagreements. Group I  $- {}^{90}$ Sr (factor 4),  132 Xe (factor 20), Group II  $- {}^{103}$ Ru (factor 20), Group III  $- {}^{128}$ Te (factor 6),  144 Nd (factor 100),  148 Nd (factor 7)  150 Nd (factor 3) Group IV  $- {}^{82}$ Se (factor 10),  110 Pd (factor 10),  133 Xe (factor 30)

disagreement : in the case of ¹⁴⁶Nd for example the Japanese adopt obsolete values dating from 1962 and 1966 and seem to neglect the resonance parameters and an experimental value dating from 1967.

A reference and data compilation problem also arises : for example the Japanese seem to base their evaluation for  132 Xe on an experimental result (RI_c = 0.106 b) and hence adopt a very low RI_c value.

A comparison of available experimental data should help to resolve this disagreement.

# 4.3. Fast range.

This field may be characterised by the value of  $\sigma_e$  (30 keV) and the integral of  $\sigma_e$  over a fission spectrum (FSA). The disagreements are so large that we shall merely list the nuclei which fail to satisfy the precision requirements (table VI).

The required precision is only obtained for certain monoisotopic natural elements for wich many experimental measurements exist, and for nuclei with low cross-sections. The discord is partly due to the fact that almost all the cross-sections are obtained by calculation, even when experimental data exist, which sometimes destroys the agreement.

We have to be careful about the credibility of experimental data. There are some for ¹¹ nuclei out of the 28 selected by the Japanese working Group [Ja 73, Ma 73]; there are several sets of experimental data wich overlap between the energy ranges for 3 of them:  103 Rh,  109 Ag and  133 Cs. In his contribution MATSUNOBU [Ma 73]notes:" There are some discrebancies between the experimental data. Especially for  103 Rh  109 Ag and  133 Cs it is noted that considerable discrepancies exist between the experimental data". One would be tempted to conclude that the only way to obtain acreement between experimental data is to have only one experimental result...

We should not be so pessimistic but is has to be recognize that most of the capture experimental data above a few keV are unreliable within 20%.

Before tackling the problems of error sources and parameters of the statistical model a last comment is necessary. The disagregments have a strong systematic commonant : at 30 keV for example the Japanese and Italian values are almost always higher than those of SCHENTER (by about 60% and 25% respectively). The systematic difference depends on mass number and energy : at 30 keV the Australian values (COOK) are 20% higher than the Italian (BENZI) for  $A \leq 110$ , but for  $A \geq 130$  thev are lower. On the other hand it appears from DEAN'S calculations [DE 73] (integral of q (E) over a fast reactor spectrum) that whatever the mass number, 3/4 of the integrated Italian values are higher than the Australian values, the average ratio being about 1.5.

## Table VI

Nuclei not meeting precision requirements on  $\sigma_e$  at 30 keV and on the integral of  $\sigma_e$  (E) over a fission spectrum (FSA).

Group I	93 _{Zr} , 96 _{Zr} , 97 _{Mo} , 98 _{Mo} , 99 _{Tc} , ( ¹⁰⁰ _{Mo} ), ¹⁰¹ _{Ru} , ¹⁰² _{Ru} , ¹⁰⁴ _{Ru} , ¹⁰⁵ _{Pd} , ¹³² Xe, ( ^{13.} Cs), ¹³⁴ Xe, ^{13.} Cs ¹⁴³ Nd, ( ¹⁴⁰ Ce)
Group 2	$(\frac{95}{145}_{\text{Nd.}})$ , $\frac{103}{\text{Ru}}$ , $(^{103}_{\text{Rh}})$ , $\frac{106}{\text{Ru}}$ , $^{107}_{\text{Pd}}$ , $^{108}_{\text{Pd}}$ , $(^{131}_{\text{Xe}})$
Group 3	$\binom{83}{\text{Kr}}$ , $\binom{85}{\text{Rb}}$ , $\binom{109}{\text{Ag}}$ , $\binom{129}{\text{I}}$ , $\frac{144}{\text{Nd}}$ , $\frac{147}{\text{Pm}}$ , $\frac{149}{\text{Sm}}$
Group 4	¹⁵¹ Sm
Group 5	$(\frac{147}{\text{Sm}})$ , $(\frac{153}{\text{Eu}})$ , $\frac{155}{\text{Eu}}$

Note : Nuclei of half-life shorter than 30 years are underlined. Those which do not satisfy only one of the 2 criteria are bracketed.

#### 5. ANALYSIS OF ERROR SOURCES.

Two contributions [Sc 72, Dr 73] examine the source of errors in the calculated cross-sections. In the first, SCHMITTROTH takes into consideration only the influence of the statistical resonance parameter fluctuations : he gives formulae allowing to calculate the uncertainty as a function of the energy range considered L, the mean spacing D of resonances.

This is a large source of error, especially when the mean number of resonances n = L/D is small, but the error is random and should cancel out on an average. The main point is that it is preferable to describe cross section by the resonance parameters when n is small.

Many sources of error exist, sometimes due to deliberate approximations (weighting flux spectrum when calculating multigroup cross-sections). However the Dutch studies [Dr 73, La 73] demonstrate the importance of the different statistical model parameters :  $\Gamma\gamma$  and Dobs are the main sources of error on  $\sigma_c$  beyond  $\sim 100$  eV. By way of example, figure 1, taken from [Dr 73], shows the different errors in the case of 101Ru and  $108_{Pd}$ .

It should be noted that these studies assume the validity of the statistical model : certain phenomena such as a correlation between  $\Gamma\gamma$  and  $\Gamma n$  would contribute systematically to the error.

## 6. SYSTEMATICS

This involves collecting all the parameters from which the description of the nucleus is obtained, so as to derive laws describing their variations. It can apply either to quantities directly obtainable by experiment (Dobs,  $\overline{\Gamma}\gamma$ ,...) or to the parameters of a theoretical model (optical model parameters, the level density parameter a, etc...).



Fig. 1. Contributions to the standard deviations of the group cross sections for the 26 ABBN energy groups for the isotopes ¹⁰¹Ru and ¹⁰⁸Pd (Taken from ref. [Dr 73]) Explanation of symbols:

```
+ = resolved resonances ; x = 1/v contribution;

Δ = photon strength function; □ = s-wave strength function;

Φ = p-wave strength function; ∇ = d-wave strength function.

* = statistics of levels ;

Φ = sum of contributions listed above;

• = total error, including systematic and statistic model-errors
```

It is not possible here to examine all recent studies. One or two only will be mentioned, after which some problems relative to level spacings will be discussed.

## 6.1. Optical model parameters and strength functions.

It is worth mentioning the compilation of  $S_0$  and  $S_1$  by A.R. de L. MUSGROVE [Mu 73] and that of PEREY on the optical model parameters [Pe 72] (note 1).

Mac KELLAR and SCHENTER [Ma 72] study the influence of the optical model parameter values on  $\sigma_i$  and note that it is particularly strong for  $E \leq 10$  keV, at 1 keV for example ; various realistic potentials give variations of a factor 2. The same effect was observed at Saclay and it was concluded that for preferance the strength functions should be used below about 30 keV.

## 6.2. Statistical model parameters.

There are essentially the radiative width  $\overline{\Gamma}\gamma$ , the average spacing Dobs and the level density parameter a. The first two parameters can be obtained directly by experiment and a is usually deduced from the spacing Dobs, or in some cases from the form of the evaporation spectrum of certain reactions. Conversely the theoretical estimation of  $\overline{\Gamma}\gamma$  and Dobs often used for F.P.'s is based on the knowledge of the parameter a.

# 6.2.1. Experimental and theoretical determination of a.

Two processes are used to predict the a value as a function of the number of nucleons of the nucleus.

- a) by a graphical method a is plotted against the mass number, or better still against the number of neutrons using experi-
- Note 1 : This 1972 study is being brought up to date, particularly for the neutron potentials.

TABLE VII - MEAN LEVEL SPACINGS D (eV) AND LEVEL DENSITY PARAMETERS a (Mev⁻¹)

Compound	FACCI [FAC	HINI 58]	BAE [BAT	*A 70]	BENZ BET	2 I 9]	SCHMIT [SC]	TRUTH 73 <b>]</b>	WEIGN (WET	ANN 3]
nuclei	D _{obs}	8	D _{obs}	8	D _{obs}	8	Dobs	a	Dobs	8
72GA 73GE 74GE	1.65 E2 2.200E3 7.700E1	13.37 13.39 13.07	1.9 E2 3.900E3 7.700E1	12.76 12.10 12.86	3.810E2 1.550E3 1.240±2	11.90 13.75 12.55	1.4 E2 1.787E3 7.480E1	12.57 12.48 12.52	1.550E3 6.200E1	10.00 10.60
75GE 77GE 76AS 75SE	8.000E3 8.000E1	12.80 13.19	8.500E3 5.000E3 8.730E1	11.58 12.54 12.81	5.850E3 4.200E3 7.140E1	12.30 13.9 13.10	4.987E3 5.430E3 7.540E1 3.53 E2	11.34 12.16 12.29 13.02	3.900E3 4.200E3 7.500E1 3.70 E2	9.25 9.80 10.30
77SE 78SE 79SE	1.450E3 1.100E2 3.700E3	13.06 13.29 12.38	1.200±3 1.500±2 4.500±3	13.05 12.65 12.00	9.330E2 1.200E2 1.000E3	14. 13.40 14.34	9.540E2 8.980E1 2.540E3	12.55 12.54 11.79	7.000E2 1.200E2 1.000E3	10.20 9.85 10.30
83SE 80BR 82BR	4.350E3 7.05 E3 5.700E1	12.60	6.90 E3 6.100E1 5.200E1	14.05 13.25 12.69 13.33	4.110E3 6.000E1	13.35	2.90283 9.13683 6.31081 1.03082	12.09 11.57 11.83 11.56	6.000EL	9.99
8688 8888 8558 8758	6.200E2 1.700E3 3.500E2	8.72 10.69 13.26 10.98	1.100E3 1.600E3 3.500E2 2.100E3	8.66 10.98 13.17 11.09	1.800E2	10.65	3.080E2 5.030E2 4.400E2 1.975E3	8.81 11.52 11.85 9.87		
885R 895R 90 Y	2.300E2 1.000E4 2.000E3	9.36 9.25 8.64	2.100E2 1.200E4 1.600E3	9.82 13.47 10.17			3.240E2 4.701E4 3.476E3	8.52 6.00 8.67	2.000E3	8.81
91ZR 92ZR 93ZR 95ZR	5.000E3 2.300E2 3.300E3 2.500E3	11.20 12.39 12.78 13.90	3.300E3 2.500E2 3.400E3 3.300E3	12.22 11.62 12.95 13.57	5.690E3 3.360E2 3.560E3 1.810E3	11.70 13.20 14.42	5.070E2 2.300E3 5.689E3	10.12 10.34 12.21 11.25	5.300E3 2.400E2 1.090E3 2.330E3	9.59 10.30 11.30 10.30
97ZR 94N8 9340 9540	2.400E3 3.6 E1	16.47 12.46	1.100E3 3.6 E1	17.63 13.15			1-13683 8.97 E1	16.56 12.03	7.500E2 3.25 E1 2.8 E3 1.8 E3	13.50 12.40 8.79 10.20
96MD 97MD 98MD 98MD	5.700E1 6.000E2 8.000E1 2.300E2	12.87 16.03 15.45	1.000E2 1.200E3 1.200E2 7.900E2	12.87 14.69 14.71	5.100E1 1.200E3 5.000E1 9.400E2	15. 14.60 15.94 18.42	1.140c2 1.367E3 7.750E1	12.49 13.17 14.39 16.04	1.020E2 1.200E3 8.000E1 9.400E2	10.80 11.10 11.50 12.90
101MU 100TC 100RU	3.000E2 2.100E1	19.28	4.000E2 2.600E1 2.000E2	18.56 15.72 12.46	7.700E2 2.440E1 3.400E1	19.50 15.60 14.96	1.339E3 3.460E1	17.14	7.700E2 2.440E1 3.800E1	14.00 13.10 11.20
102RU 103RU 105RU 104RH	2.800E1	15.34	1.030E1	10.22	3.380E1	15.60	2.85 E2 2.740c1	14.98 18.73 15.81	2.500E1 5.4 E2 2.8 E2 2.700E1	12.50 13.50 13.00
106PD 108AG 110AG 112CD	1.330E1 3.500E1 1.5 E1 3.300E1	16.71 16.12 18.01 17.06	1.110E1 5.000E1 1.91 E1 3.400E1	16.86 15.61 17.93 16.97	1.750E1 1.9 E1	17.10 18.2	1.010E1 3.220E1 1.95 E1 3.240E1	16.07 15.08 16.65 15.86	1.350E1 1.28 E1 2.600E1	13.50 14.30 12.90
113CD 114CD 115CD	2.700E1	18.74	2.000E2 2.700E1	19.20 18.51	1 10051	15 90	2.5201	17.53	1.980E2 2.450E1 1.57 E2	13.60 13.40 14.90
1141N 116IN 113SN 115SN 116SN	0.5 6.5 4.000E1	10.77 18.11 18.79	7-1 9-5 1-400E2 3-200E2 5-000E1	17.42 16.57 15.78 16.24	1.000E1	17-10	1.160E1	16.21	1.000E1	13.70
1175N 1185N 1195N 1205N	1.800E2 4.500E1 3.200E2 5.200E1	18.15 10.43 18.37 16.88	2.500E2 6.500E1 7.300E2 6.200E1	17.49 16.03 15.08 16.74	2.500E1	17.30	5.720E1 5.140E2 1.790E2	14-63 15-60 13-60	2.490E2 2.500E1 5.020E2	14.00 14.20 13.30
121SN 123SN 125SN 122SB	2.500E2 4.000E2 2.500E2 1.200E1	17.00 19.81 18.42 17.47	2.400E2 4.000E2 2.500E2 1.300E1	17.74 17.57 19.01 17.43	1.000E1	17.40	7.44062 4.88613 1.37011	16.06 13.06 16.10	4.060E2 1.320E3 1.360E3 1.000E1	15.10 13.90 14.90 14.70

## FROM DIFFERENTS AUTHORS.

•

Compound	FACCI [FA	HINI 68]	BAI [BA	8A 70]	BEN [BE7	Z I 3]	SCHMIT [SC	TROTH 73]	WEIG [WE	MANN ' 73]
nuclei	D _{obs}	a	D _{obs}	8	D _{obs}	a	0 _{obs}	a	Dobs	a
124SB	2.90011	16.44	3.000E1	16.62	2.100E1	17.00	2.340E1	15.78	2.100E1	14.40
I23TE	1.150E2	18.99	1.300E2	18.68			1.320E2	17.27	1.320E2	14-40
124TE	2.200E1	18.10	3.300E1	17.51	2.920E1	17.60	2.630E1	16.51	2.630E1	13-60
125TE	2.100E2	18-81					1.470E2	18.05	1.470E2	15.20
126TE	6-000E1	16.32	4.000E1	17.02	3.780E1	16.80	3.780E1	15.87	3.780E1	13.70
127TE							2.070E2	18.25	2.070E2	15.70
129TE	5.500E2	18.38	5.500E2	18.51			2.630E2	18.41	2.620E2	15.90
131TE	3.500E3	15.08	5.700E3	15.03			8.720E2	16.59	8.720E2	15.50
128 I	1.520E1	17.13	1.900E1	17.02	1		1.470E1	16.06	1.300E1	14+40
130 I	2.70051	10.49	2.100E1	17.20			2.610E1	15.52		₹ ⁴
130XE			-				3.62 E1	15.84		
132XE			3-100E1	10.97			3.92011	12.02		
136XE	2 05051	14 64	5.000E2	14.55	1.020(1	14 (0	2 02051	16 66	2'00051	12 00
13465	2+05051	10+20	2.01021	10+11	1.92021	10.40	2.020EL	11 90	2.00051	13.74
12104	1 20052	19.22	1 7 62	18 47			rett CT	11.00	• .	
13584	3.80052	17.37	1.8 E2	17.69					1.30052	15.10
13684	3.70051	15.98	3.5 61	16.56	2.600F1	17.00	3.71051	14.81	2-60051	13.50
13784	3-500E2	18.23	3.8 63	14.56	2.000011	11000	Jerroun		2000022	
13884	4-00052	13.71	4.6 F2	14.35		}	5-220F2	12.14		, i
13984	TROUVLE	12011	9060 E3	19.33			J			
13914	3-10 F1	13.46	4.10 F1	13.76	Ì		3-570F1	12.58		
140LA	9.00 E1	18.36	1.10 E2	18.29			3.120E2	14.68	1.00 E2	16.20
141CE			3.00 E3	17.80						
143CE			1.00 E3	21.31						
142PR	6-40 E1	16.99	8.38 E1	17.05	3.120E1	18.25	6.390E1	15.77	7.50 E1	14.70
143ND			1	•			4-150E2	17.83	4-150E2	16.00
144ND	2.600E1	18.24	1.900E1	19.15	3.200E1	18.00	3.200E1	16.70	3.200E1	15.30
145ND	{				5.2 E2	20.21	5.37 E2	18.48	5.37 E2	15.60
146ND	2.000E1	20.09	2.500E1	19.87	1.770E1	20.40	1.890E1	18.94	1.890E1	16.50
147ND				1	3-100E2	23.54	2.110E2	22.59	3.100E2	18.30
149ND			1		2.580E2	26.00	7.200E1	26.81		税
151ND					2.470E2	24.90		23.57		
148PM	4.0	21.98	5.7	21.30	4.76	21.80			4.10	18+40
1485M		20.36	1.7	20.50	1.4	20.51	8.18	19.01	1.4	10+10
15054	<.+	23.01	3.44	25 49	2.0	23.30	4.08	21.00	4 90051	20 70
15204	1.	26.64	1.3	24.70		l			0.0UVEI	20010
1525M	1.	22.20	1	24.10	5 35051	25. 40			5 20 E1	10.20
1555M		}	U. CI	67071	1.25052	24.60			1.25052	17.70
152FU	0.65	25.05	0.72	24.58	1.	24-10			1.04	20 - 30
154FU	1.250	23.36	1.30	23-24	1.45	22.80			1.45	19.40
153GD					1.9 F1	24.6		1	1.9 E1	19.70
155GD		ļ		ł	1.9 E1	24.7			1.9 E1	19.80
156GD	1.9	22.45	1.99	22.46	1.98	22.20			1.99	18.20
157GD	7. El	21.94	7.50 E1	21.98	4.930E1	22.80		1	5. El	17.60
158GD	5.5	21.60	6.1	21.64	5.85	21.40			5.0	17.20
159GD	1.7 E2	21.15			1.010E2	22.90		1	8.400E1	18.30
16160		4		1	1.700E2	23.50		1	1.60062	17.90

mental values. This is the method used by Benzi et al [Be 71, Be 73]. Table VII includes a selection from the most recent Dobs and a values for a hundred or so nuclei in the F.P. mass range. The fluctuations observed on the a value can be due to two factors :

- The first is the value of Dobs which varies from one study to another.Even for a well-known nucleus, ¹⁰³Rh, the difference within the 5 groups of values reaches a factor 3.
- The second is a systematic component due to differences between the formulae and values used in particular for the spin cut-off factor and the effective excitation energy.

Table VIII shows the variation in the mean a values taken within 4 mass ranges.

This table shows up clearly a systematic divergence ascribable to the different versions of the level density formula. The expressions used by the authors whose values appear in table VII are given in the appendix.

b) By adjustment of theoretical formulae. Weigmann and Rohr
 [We 73] and Schmittroth [Sc 73] have introduced certain coefficients into the theoretical expressions to account

## TABLE VIII

								-		
	FACCHINI		BABA	BABA		BENZI		TROTH	WEIGMANN	
Authors	[Fa	68]	[Ba	70]	[Ве	ə 73]	[Sc	73]	[We	73]
	a	Da	a	Da	a	Da	a	Da	a	Da
average over 9 nuclei A ≤ 47	13.0	0	12.8	-0,2	13.6	+0,6	12,3	-0,7	10.2	-2,8
average over 10 nuclei 52 < A < 62	15.5	0	15.2	-0,3	16.1	+0,6	14.3	-1,2	12.1	-3,4
average over 7 nuclei 63 ≼ A ≼ 72	17.1	0	17.0	-0,1	17.3	+0,2	15.85	-1,3	11.9	-5,2
average over 4 nuclei 84 < A < 87	18.9	0	19.2	+0,3	19.3	+0,4	17.5	-1,4	15.8	-3,1

Mean a values according to different authors and divergences with regard to those of reference [Fa 68]. for the experimental variation of a. Thus the latter author, whose formalism is based on the model of the independent particle in a spherical nucleus, has added about twenty parameters.

# 6.2.2. Calculation of Dobs and $\Gamma\gamma$ .

Schmittroth [Sc 73] has compared, for 84 nuclei, the experimental data of Dobs to calculated values using Dobs values with the theoretical a value. The average disagreement reaches a factor 1.87, which means that it is not yet possible to predict the mean spacing of any nucleus to better than a factor 2. Two contributions deal with the mean radiative widths  $\Gamma\gamma$ . Weigmann and Rohr [We 73] introduce a direct reaction component, while Benzi and al. [Be73] compare the three formalisms of Brink, Weigmann and Rohr, and Musgrove. At binding energy the three estimates of  $\Gamma\gamma$  are equally acceptable and in one case out of two lie within the limits of experimental error, often very large. This is no longer true when the energy varies and it would then be preferable to use a formula derived from Brink's model supplemented by a direct interaction term.

# 7. OTHER CROSS-SECTIONS. SECONDARY SPECTRA.

Very few demands appear in the literature for data other than the capture cross-section (note 1). We shall nevertheless deal specially with the case of inelastic scattering since this is the most important cross-section after that of capture, and since several studies already exist.

# 7.1. Inelastic cross-sections.

All the Australian estimates contain the values of this cross-section whereas in the Italian library it is only given for 11 nuclei. Figure 2 shows the total inelastic cross-section in the case of  151  Sm.

Note 1 : This remark was confirmed by the Panel discussions.



Fig. 2 - Total inelastic cross section of ¹⁵¹Sm. At Saclay, the results were obtained with the code FISINGA and a new evaluated level scheme.

----

This is the most extreme case observed, but disagreements at about 500 keV exceed a factor 2 in half the cases. This is due to the lack of effort so far devoted to the study of inelastic scattering by fission products.

The Australian library has used a very simple model : for all nuclei the inelastic scattering threshold is at 0.5, 1 or 2 MeV and  $\sigma_{in}$  a regularly increasing curve, independently of any level scheme.

In fact the work carried out at Saclay has shown that inelastic cross-sections can be calculated to  $\sim$  30-40% (in the standard deviation sense) as long as the level scheme of the nucleus is known. Other authors reach the same conclusion [Gr 73 a, Ja 73].

If the scheme of the levels excitable by inelastic neutron scattering is unknown it is impossible to predict, even to within a factor 10, the inelastic scattering crosssection.

Conversely it seems that the scattering cross-sections cannot be calculated to 20% or better at present because of both the limitations of the optical models used and a considerable uncertainty over the parameters to be employed ; in the case of  103 Rh, for which the inelastic cross section below 300 keV, is well known, we were unable to obtain an agreement to within 20%.

The use of more refined optical models (coupled channel models) should improve the precision, and tests are in progress along these lines for the fourth version of  $E \ N \ D \ F \ / B$ .

However a 30 to 40% precision, obtainable by a spherical optical model with good parameters and with an accurate or realistic level scheme, should be enough for fission products (note 1).

Note 1 : During the Panel discussion the precision required was given as around 50%

#### 7.2. Other cross-sections.

No mention is made of the accuracies desired and hardly any theoretical evaluations exist. Experimental data in reasonable quantities exist only for monoisotopic stable fission products ( 103 Rh,  127 I, etc).

Given the low precision likely to be required it is safe to assume that any needs could be met by evaluations without recourse to experimental measurements as long as the threshold energies of reactions such as (n, 2n), (n,p) etc... are known, as is generally the case.

## 7.3. Secondary spectra.

There again no demand appears (note 2), although it is obvious that a knowledge of the inelastically scattered neutron spectrum is nearly as important as that of the  $\sigma_{in}$  value. The spectrum is not given by the Australians but appears in E N D F and for 11 F.P.'s in the Italian library.

A temperature law is very open to criticism, especially at low incident neutron energies [LH 72]; once again it is only by knowing the level scheme that a satisfactory knowledge of this spectrum can be obtained. Beyond 1 MeV a temperature law, though far from perfect, is certainly adequate as long as an adjusted value of T is used, i.e. a value corresponding to an exact average of the secondery neutron energy.

# 7.4. y rays.

No evaluations exist in the libraries, though we should mention a semi-empirical method of HOWERTON and PLECHATY [Ho68] by which the spectrum of  $\gamma$  rays produced by interaction of high-energy neutrons ( $\geq$  4 MeV) may be determined approximate-ly.

Note 2 : The small importance of these data was confirmed during the Panel discussion.

8. EXPERIMENTAL ACTIVITIES.

As shown above, the only data requested are capture cross-sections.

## 8.1. Capture cross-sections.

We have only examined the case of nuclei belonging to group 1 of table II, or appearing in the first parts of tables IV and V, i.e. 48 isotopes altogether. Experimental activities only exist for fifteen of these isotopes (table IX) and are mainly concentrated in a few laboratories : ORNL, CHALK RIVER and ARGENTINE, except for natural monoisotopic elements :  $133_{CS}$ .  $139_{La}$ ...

Several other laboratories have integral cross-section measurement programmes [Bu 73].

# Table IX

Survey on measurements recently performed in different countries

LABORATORY	DATA	NUCLEUS			
O R N L (U.S.A.)	<b>d</b> _c 3 keV to 600 keV	88 90 94 Sr, Zr, Zr, ¹⁴³ Nd,			
C N E A (ARGENTINA)	RIc	${}^{96}_{Zr}$ , ${}^{100}_{MO}$ , ${}^{102}_{Ru}$ , ${}^{104}_{Ru}$ , ${}^{138}_{Ba}$ , ${}^{140}_{Ce}$ , ${}^{146}_{Nd}$ , ${}^{148}_{Nd}$ , ${}^{150}_{Nd}$ .			
KJELLER (NORWAY)	RIc	¹⁴⁰ Ce.			
DUBNA, BORDEAUX, WUERENLINGER, MTR, KJELLER.	Several data	¹³³ cs, ¹³⁹ La.			
CHALK RIVER (CANADA)	Øact (th)	147 _{Pm} , 149 _{Pm} , ¹⁵⁵ Eu.			
STUDSVIK (SWEDEN)	RI _C	149 _{Sm} ,			

### 8.2. Other cross-sections.

Because of the lack of requests the experimental activities have other purposes : nuclear physics, structural materials (Mo), dosimetry ( 103 Rh). It is worth emphasizing the benefit to systematics of experiments motivated by no immediate need for example the study of resonance parameters which are gradually improving our knowledge of the behaviour of  $\Gamma\gamma$ , Dobs versus A. (Note 1).

# 9. CONCLUSION

For thermal reactors where the number of important F P is restricted to about thirty nuclei, the comparison between the requested accuracy and that obtained in measurements and evaluations shows that the capture data must be improved for a dozen nuclei (table IV, 1). In the fast energy range the situation requires more works both in the experimental as in the theoretical field. The knowledge of the capture must be improved for about thirty isotopes, two thirds of them have a contribution greater than 1 % of the reactivity effect due to the overall fission product mixture. (table VI). It would be usefull to undertake high precision measurements in the keV region with natural monoisotopic elements for which the available data are too discrepant.

More studies should be devoted to inelastic scattering which has been up to now neglected. In most cases a statistical model calculation suffices, in so far as the level schemes are known up to 1 or 1.5 MeV.

Note 1 : Spin measurements such as that of ASAMI for 107 and 109 Ag [AS 73] will lead to a better determination of partial widths and hence of  $\Gamma\gamma$ , but this sideline will not be available for several years.

#### APPENDIX

Formulae used to determine the level density parameter a .

A) - The various expressions of the level density formula and other related quantities.
 I - The level density (U,J)

Pormula (1) :  $p(U,J) = \frac{2J+1}{24\sqrt{2}a^{1/4}\sigma^3} = \frac{\exp(2\sqrt{aU-J(J+1)/2\sigma^2})}{U^{5/4}}$ 

Formula (2):  $p(U,J) = \frac{2 J + 1}{24 \sqrt{2} a^{1/4} \sigma^3} = \frac{\exp(2\sqrt{aU-J(J+1)/2}\sigma^2)}{U^{5/4} (1+1/\sqrt{Ua})^2}$ 

II - The effective excitation energy U Formula (3) : U = E -  $\Delta$ Formula (4) : U = E -  $\Delta$ +? A : pairing energy  $\gamma$  : shell model contribution

III - The spin cut-off factor  $\sigma$ Formula (5):  $\sigma^2 = 0.0887 \ A^{2/3} \sqrt{aU}$ Formula (6):  $\sigma^2 = 0.146 \ A^{2/3} \sqrt{aU}$ Formula (7):  $\sigma^2 = 0.06 \ A^{7/6}$ Formula (8):  $\sigma^2 = \frac{T_R}{\hbar^2} \sqrt{\frac{U}{a}} \simeq 0.177 \ A^{2/3} \sqrt{aU}$ T_R is the nuclear moment of inertia

B) - The table below summarizes the procedures used by the authors o table VII to calculate the level density parameter a.

	Réf.	p(U,J)	υ	5 ²	Bn	Δ	7
U. FACCHINI E.SAETTA-MENICHELLA	Fa 68	(1)	(3)	(6)	Wa 65	Ca 65	
H. BABA, S. BABA H. BABA	Ba 69 Ba 70	(1,2) (1)	(3)	(8) (8)	Wa 65	Ca 65	
V. BENZI et al.	Be 73	(1)	(3)	(6)			
F. SCHMITTROTH	Sc 73	(1)	(3)	(5)		Ca 65	
H. WEIGNANN & G. ROHR	We 73	(1)	(4)	(7)	Wa 71	Ne 62	Ka69

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## STATUS OF FISSION PRODUCT YIELD DATA FOR THERMAL REACTORS

## W.H. Walker

## Atomic Energy of Canada Limited Chalk River, Ontario, Canada

#### 1. SUMMARY

Fission products affect reactors in many ways. Calculations of all these effects depend on fission product yields and hence on their accuracy. The dependence on accuracy may be high, as in the case of neutron absorption in ¹³⁵Xe and fuel burnup determinations [1] or low, as in the case of total energy release [2] [3] [4] or the average  $\beta$ -decay energy per fission.

Since the purpose of this meeting is to assess the current status of fission product nuclear data from the practical rather than fundamental scientific viewpoint, the end product of this review is a recommended set of chain yields for thermal fission of ²³³U, ²³⁵U, ²³⁹Pu and ²⁴¹Pu, and an assessment of their uncertainties. Measured direct yields are not numerous enough to permit such a set. The main problem areas for both chain and direct yield measurements are noted.

#### 1.1 Contents of Review

The bulk of this paper will discuss the data and evaluation procedures on which the recommended values are based. This is done in the context of a brief historical review of yield measurements and their development. The advantages of each type of yield measurement and their sources of systematic errors are discussed.

A comparison of yield values indicates that many data do include systematic errors and it is essential that these systematic errors be taken into account. In only a few cases can errors be assigned to a particular yield value, which may then be corrected or rejected. An evaluation procedure is proposed that will enhance the probability of recognizing the presence of unidentified systematic errors and reduce the probability of assigning too great a weight to a yield containing a large systematic error.

Five recent evaluations of cumulative yields [5] [6] [7] [8] [9] are compared to the proposed procedure. On the basis of a comparison of their recommended yields and data on which they are based, sets of cumulative yields for the thermal neutron fission of  $^{233}U$ ,  $^{235}U$ ,  $^{234}Pu$  and  $^{241}Pu$  are recommended.

Information on direct yields is sparse as compared to that for cumulative yields. For ²³⁵U these have been reviewed recently by Wahl [10], Denschlag [11] and Amiel and Feldstein [12], primarily to establish the parameters of a semi-empirical model of charge dispersion.

In this paper only direct yields of interest in fuel burnup and decay heating calculations are considered. It is apparent many more measurements are required before we can dispense with a semi-empirical model of charge dispersion to predict direct yields, particularly in ²³³U, ²³⁹Pu and ²⁴¹Pu thermal fission.

Finally the few data on cumulative yields from fission by epithermal neutrons are listed and their significance is discussed briefly.

#### 1.2 Yield Compilations and Evaluations

The remainder of this summary is intended to give the "state of the art" in compiling and evaluating and the status of measurements of chain yields, fractional yields and epithermal yields.

In compiling yield measurements for evaluation the most obvious problem is the large number to be dealt with. For example, Meek and Rider [9] list over 17000 data cards, of which about 3000 are used in calculations of cumulative and direct yields from thermal fission. Some of these data have been re-examined recently, but much similar work is required before they can all be certified as valid.

Five evaluations of cumulative yields have been published in the last two years, culminating preparatory work that in some cases began nearly ten years ago. The number is remarkable in comparison to the average rate of publication of such evaluations in the preceding two decades.

The problem presented by the large amount of yield data has been tackled in two ways. Meek and Rider [5] [9] and Crouch [6] record all their data in a computer file after assessing the accuracy of each value, and then evaluate the data by computer on a mass-by-mass basis. Walker [7] [13] and Lammer and Eder [8] concentrate on mass spectrometric measurements to establish, with relatively high precision, the dependence of yield on mass for about 90% of the yield of a particular fissile nuclide, and use radiometric data mainly to fill in the remaining 10%.

This concentration of effort on yield evaluation has not been wasteful duplication because each has contributed something significantly different in procedure or insight into the sources of disagreement. Even more valuable, from the viewpoint of this meeting, is the extensive weeding out of the inevitable errors which are more easily located when one set is compared to another. This is a process which would otherwise take many years and might never be done satisfactorily. Yields of ²³⁵U fission products are much the best known. Not only have they been measured more frequently, but there are fewer disagreements, perhaps an indication that the work was done more carefully.

For reactor calculations the most serious uncertainties are in the yields of the Sm isotopes. These result from a 15% spread in mass spectrometric measurements of the atom ratios of fission product Sm and Nd.

Yields from ²³⁵U fission are used frequently as reference standards. They would be more reliable for this purpose if the yields so far neglected were measured, particularly in the valley between light and heavy mass peaks of the yield curve, and if the remaining disagreements were resolved.

Thermal cumulative yields for  $^{233}U$  are acceptable. The lack of serious disagreements however, may be merely a reflection of the scarcity of measurements.

For both ²³⁹Pu and ²⁴¹Pu additional measurements are required. These include the ¹⁰³Ru and ¹⁰⁶Ru yields, the Kr/Xe ratio and the Cs yields. New data are required for the Pd-In range, particularly since the yields are much higher from fissile Pu than from fissile U.

If the ²³⁵U yields are established more firmly by new measurements, then relative yields measurements using Ge (Li) gamma detectors can provide a rapid and reliable method of determining additional ²³³U, ²³⁹Pu and ²⁴¹Pu thermal yields, as well as fast neutron yields.

#### 1.4 Status of Direct Yield Measurements

For fission product heating calculations, direct or cumulative yields of 3 or 4 isobars are required for each mass. Even if this requirement is restricted to masses with yields exceeding 1%, nearly twice as many direct yields are required as there are chain yields.

For ²³⁵U there are cumulative or direct yields for about 70% of the nuclides in this category, but even here, for a substantial fraction, either there are significant disagreements or only one measurement has been made.

For ²³³U, ²³⁹Pu and ²⁴¹Pu very little data exists and calculations will have to rely on a set of direct yields based on a semi-empirical model fitted to ²³⁵U data.

#### 1.5 Status of Epithermal Fission Yield Measurements

The peak-to-valley yield ratio is a measure of symmetric to asymmetric fission. Using this ratio it has been established that yields from the first resonance at 0.3 eV in ²³⁹Pu are much more strongly asymmetric than for thermal fission, i.e. the yields in the valley between peaks in the mass yield curve are much reduced. Changes have also been observed for ²³⁵U and ²³³U fission, but are much smaller. The effect of these changes on reactor calculations will be negligible if only the very small yields change appreciably, but changes in large yields near the peaks are also possible. At present, the best experimental evidence available indicates that such changes are 3% or less.

In some reactor calculations epithermal yields are found by interpolation between thermal yields and reactor spectrum yields. Since the latter are enhanced in the symmetric fission mode, such interpolations could lead to small errors if they are applied below about 100 eV.

#### 2. FISSION PRODUCT YIELD MEASUREMENTS

#### 2.1 A Brief History

The first yield measurements were made by Hahn and Strassman [14] when they detected the presence of radioactive Ba and thus established that their uranium target was fissioning rather than capturing when bombarded with slow neutrons.

Soon after that it became apparent that fission into fragments of approximately equal mass had a small probability, and the first phase in yield measurements was concerned with establishing quantitatively the double-humped dependence of yields on fission product mass.

These early distributions had many disagreements and certainly did not give a smooth variation of yield with mass. Initially wide deviations from a smooth curve were attributed, with good cause, to uncertainties in the measured yields, but, as techniques were refined, some anomalies persisted. These became known as "fine structure". In the second phase, beginning in the early 1950's, the main interest in yield measurements was in determining the location and magnitude of fine structure, and explaining it in terms of the shell model of the nucleus.

By the early 1960's the majority of chain yields in  ${}^{235}U$ ,  ${}^{233}U$  and  ${}^{239}Pu$  fission had been measured. In the third phase interest, particularly among radiochemists, turned to measuring direct yields of fission products and their isobaric distribution immediately following neutron emission, and in testing several hypotheses intended to predict that distribution.

In the mid-1960's, as more and more power reactors came into operation, it became important to determine fuel burnup not only for assessing reactor performance, but also for fuel processing and safeguard inventories. Fission products can be used for this purpose if their yields are known accurately. Several nuclear energy laboratories were active in this fourth phase of yield measurements, with the most extensive work being done at Idaho Falls [15].

During the same period Ge(Li)  $\gamma$ -ray spectrometry was used to determine the yields of radioactive fission products having half-lives of about an hour or longer. More recently these detectors have been used to measure yields of shortlived fission products including direct and cumulative fractional yields [90]. These measurements should have their greatest application for ²³³U and ²³⁹Pu fission since the direct yields of longer-lived nuclides is expected to be greater for these fissile nuclides than for ²³⁵U and ²⁴¹Pu.

#### 2.2 Methods of Yield Measurement

Each of the phases listed in the preceding outline was associated with the development of a particular method of measurement, or a significant improvement in an established method.

#### 2.2.1 Radiochemistry, Absolute Yields

This is the oldest method of measuring yields, the method of Hahn and Strassman. It played the major role in the first phase of yield measurements.

Briefly the fissile material is irradiated and dissolved. The solution is treated chemically to separate a particular fission product element or group of elements. The separated product is placed in a radiation detector and its counting rate is measured as a function of time. The number of fission events must also be determined.

The samples are  $\beta$ -counted except in cases where a  $\gamma$ -ray emission probability is well known. Originally thin-window or thin-wall Geiger-Müller counters were used. For later  $\beta$ -counting the sample was placed inside a gas-flow counter with high geometry. Counts from different isotopes were resolved using half-lives and  $\beta$ -spectrum end-point energies (or transmission through filters).

Because radiochemical yield measurements can be made with very small samples it is the preferred method for determining low yields such as chain yields at near-symmetric and very asymmetric fission, and direct yields.

2.2.2. Radiochemistry, Relative Yields

Once the yield of one radioactive fission product is known precisely, it can be used as an internal monitor of the number of fissions. Favorite fission products to fill this role are  $MO^{99}$ ,  $Ba^{140}$  and  $Cs^{137}$ . This procedure was used for many of the later measurements of the first phase, and is the one most widely used in the determination of charge distribution.

The advent of Ge(Li) detectors and the subsequent accurate determinations of  $\gamma$ -ray emission probabilities for many radioactive fission products has greatly improved the ease and accuracy of relative yield determinations.

## 2.2.3 Radiochemistry, R-Value Method

When ²³⁵U yields became reasonably well-known it was possible to eliminate one of the major sources of uncertainty in radiochemical measurements - those due to errors in estimating geometric losses and in counting corrections arising from uncertainties in the decay schemes. To do this, samples of two fissile atoms are irradiated together and each is processed to obtain the fission product elements to be counted. Let the subscript "r" refer to the "reference" fissile nuclide, the one for which the yields are assumed known; "u" to the "unknown" fissile nuclide for which the yields are to be determined; "s" to a "standard" radioactive fission product such as ¹⁴⁰Ba; and "x" to the fission product for which the yield in the unknown is required. If A refers to a measured activity corrected to a chosen time such as the end of the irradiation and y refers to yield, then the R-value is defined as the ratio of activity ratios given by,

$$R = \frac{A_{ux}}{A_{rx}} \frac{A_{rs}}{A_{us}}$$

The required yield is given by

$$y_{ux} = R(y_{rx} y_{us}/y_{rs})$$

Note that the yield y_{us} must be known in addition to those of the reference fissile nuclide.

This method reduces the chance of errors due to processing losses but does not eliminate them since identical treatment of different fissile materials need not lead to equal recovery of a particular element.

For a discussion of experimental techniques with references to the original work the recent review by von Gunten is recommended [16].

#### 2.2.4 Mass Spectrometry

Mass spectrometers have been used in fission product yield measurements since the mid-1940's. As in radiochemical measurements the irradiated fissile material must be dissolved and the various elements separated.

Mass spectrometers give the relative abundances of the isotopes of the element under study. These cannot be converted to yields without further measurements or assumptions. In the earlier work they were normalized to an assumed yield at a particular mass, usually based on a radiometric measurement. For adjacent elements with isotopes alternating in mass, for example ¹⁰⁴Ru, ¹⁰⁵Pd, ¹⁰⁶Ru(1 yr), ¹⁰⁷Pd, the yields might or might not vary smoothly with mass depending on the relative magnitudes of the two normalizing yields.

This uncertainty in normalization was a serious problem in measurements of fine structure. In order to improve the relative normalization of adjacent elements, the techniques of isotope dilution and isobaric coupling were perfected, notably at McMaster University. The use of these techniques is discussed in greater detail elsewhere [7] [8] [17]. Mass spectrometry provides an accurate method of determining burnup as well as fission product element yields provided the sample is irradiated long enough to change the number of fissile atoms significantly. Lisman et al [15] used this technique to determine the absolute yields from thermal neutron fission of 233U and 235U.

Mass spectrometric measurements require relatively large samples and have, until recently, been restricted to the higher yield elements, Kr through Ru and Xe through Sm.

## 2.2.5 Y-Spectrometry

The advent of solid state detectors permitted a quite new approach to radiometric yield measurements. The essential difference from radiochemical methods is that no chemistry is performed on the irradiated samples. Rather, they remain sealed during both irradiation and counting so that there is no problem of fission product losses. If the fission products are separated chemically and then counted with a Ge(Li) detector the data is treated, in this review, as radiochemical.

Background rates in the sealed samples are quite high and this limits application of the method to yields > 1% and  $\gamma$ -ray emission probabilities > 10%. Both limits can be lowered if the background is reduced using Compton detectors in anti-coincidence.

 $\gamma$ -spectrometry can be used to determine yields either absolutely [18], or with the R-value technique of section 2.2.3 [19] [20] or by comparing fission product  $\gamma$ -rays from the irradiated sample with standard sources prepared chemically from relatively large samples [21]. Of these the R-value technique appears to have the greatest potential since it can be used for fast fission yields and direct or cumulative yields of short-lived nuclides.

2.2.6 Miscellaneous Methods

4.2

The following methods have been either used in special situations or developed as a ternatives to the preceding methods, but have never been applied widely.

- (i) Volumetric determination of rare gas fission products the ratio of Xe to Kr was determined [22]. The claimed accuracy was high but the ratio disagrees with that for currently accepted yields by  $\sim 11\%$ .
- (ii) Integrated current mass spectrometry the current of ions at each mass was determined absolutely using measured ionization efficiencies for each element [23]. The method appears to have considerable potential, especially for the determination of monoisotopic fission product elements such as Tc and Pr but was never perfected. The isotopic ratios reported for multi-isotopic elements were so different from those measured by conventional mass spectrometric methods that all data must be suspect [7].
- (iii) Pile oscillator the relative yields of ¹³⁵I from ²³³U, ²³⁵U, ²³⁹Pu and ²⁴¹Pu thermal fission were

determined from the relative magnitudes of the ¹³⁵Xe absorption transient [24] [25]. The accuracy is determined by the estimated number of fissions occuring in each sample.

- (iv) Vapor phase chromatography used by Lisman et al.
   [15] to determine the number of atoms of fission product Xe and Kr.
  - (v) Spectrophotometry to determine the yield of ⁹⁹Tc [15].
- (vi) On-line mass separator a beam of fission fragments is separated according to m/e by a double-focussing mass separator and collected on photographic plates. After most of the fragments have  $\beta$ -decayed the plates are developed and the number of  $\beta$  tracks at the end of each fission fragment track are counted. From this information the charge distribution for each mass can be determined.

The method is limited to mass ranges where interference between beams with different m and e values is small. The most extensive data to date gives yields for the masses between 131 and 140 [26].

#### 2.3 Sources of Error in Yield Measurements

2.3.1 Number of Fissions

In the early radiochemical measurement of absolute yields this was one of the most serious sources of error because both flux and fission cross section were poorly known and the number of fissile atoms was difficult to determine.

In more recent radiochemical work this problem may be less serious provided measurements of well-established yields are included. Then, even if a significant error has been made, its presence can often be recognized and corrected for by treating the measurements as relative rather than absolute yields.

In mass spectrometric work the numbers of fissions have most frequently been determined, as in radiochemistry, using a flux monitor, a determination of the number of fissile atoms and an estimate of the fission cross section. The most reliable method is mass spectrometry of the fissile sample after irradiation [15]. In evaluation of mass spectrometric yields (section 3) summing the total yields to 100% in the light and heavy masses provides a reliable alternative.

## 2.3.2 Normalization of Relative Yields

The use of a standard yield not only avoids determining the number of fissions directly but also provides a straightforward method of updating the measurements as knowledge of the standard yield improves. The main sources of error are:

(i) uncertainty in the standard yield

- (ii) processing losses of the standard nuclide (the nuclide with the standard yield)
- (iii) incorrect decay data for the standard nuclide
- (iv) errors in the counting corrections for the standard nuclide.

The first can be greatly reduced if the standard nuclide has an isobar for which the yield has been determined mass spectrometrically, such as ¹⁴⁰Ba or ¹⁴⁴Ce, rather than otherwise (⁹⁹Tc, ¹⁴¹Ce). Corrections can be made for the last two errors if sufficient information on the original corrections is given.

#### 2.3.3 Processing Losses

All of the element of interest may not be recovered in the chemical extraction. In radiochemistry the amount of fission products is very small and a carrier element is usually added to facilitate extraction. Repeated extractions can be used to obtain full recovery provided the fission product is converted to the same chemical state as the carrier. In some cases losses can be monitored with a  $\gamma$ -detector.

R-value results may also be affected, especially if the two fissile atoms are isotopes of different elements such as U and Pu. In this case differences in chemistry may lead to different losses of a particular fission product element.

In mass spectrometry processing losses will not affect isotope dilution measurements provided all of the fissile sample can be dissolved.

#### 2.3.4 Contamination

Contamination with naturally occurring elements may occur in the preparation of the fissile sample, in the dissolution of the irradiated fissile sample, in the separation of a particular element during isotope dilution, or on the mass spectrometer filament.

Only the first type of contamination can affect counting measurements, but a greater range of contaminants may be troublesome since chemically similar elements may be isolated with the element of interest and interfere in the counting.

All of these sources of contamination can affect mass spectrometry and are probably the main source of error in isotope dilution measurements. In many cases, such contaminants can be recognized by the presence of isotopes not formed in fission and the necessary corrections made. Elements where this is not the case, or where such isotopes have such a low abundance in the naturally occurring element that they are very insensitive contamination monitors are Rb, Cs, Ba, and Ce.

# 2.3.5 Decay Data (Half-Lives, $\beta$ - and $\gamma$ -Energies, $\beta$ - and $\gamma$ -Emission Probabilities)

Corrections can readily be made for changes in these data if the experimenter lists all relevant input. Unfortunately most do not. All radiochemical measurements except those using R-values are subject to these errors. Some mass spectrometric yields must be corrected for  $\beta$ -decay and are therefore affected by uncertainties in the half-life involved.

#### 2.3.6 Neutron Capture

Usually, only mass spectrometric samples are irradiated sufficiently that the measured isotopic abundances need to be corrected for neutron capture. For non-saturating fission products, such as ¹³¹Xe or ¹⁴³Nd, this correction can be calculated accurately and is often negligible. For ¹³⁵Xe and ¹⁵¹Sm, the corrections can be very large and sometimes impossible to calculate accurately. ¹⁴⁹Sm differs in having as capture product a shielded isotope, ¹⁵⁰Sm, so that the sum of the ¹⁴⁹Sm and ¹⁵⁰Sm abundances is equal to the ¹⁴⁹Sm yield except for small corrections.

## 2.3.7 Counting Corrections

These are the most widespread cause of error in  $\beta$ - and  $\gamma$ -counting. They include:

- (i) Loss of radiation by absorption in the sample, air gap, and detector housing
- (ii) Backscattering of  $\beta$ 's from the mounting material
- (iii) Counting losses in the detector (end-effects, ion recombination)
  - (iv) Counting losses in the associated electronics (deadtime, pile-up)
    - (v) Interfering activities, background subtraction.

In most experiments there may be corrections for these effects, but these depend on the state of knowledge at the time of the experiment.

The accuracy of these corrections, assuming they have been made in the data, is difficult to assess at a later date. However, it is to be expected that this accuracy will improve with time. For this reason, older radiochemical data are usually assigned a larger uncertainty than more recent results.

## 2.3.8 Magnitudes of Errors

The preceding review of sources of error has been qualitative for the good reason that the magnitude of a particular type of error depends as much on the experimenter as on the method. Also, although an evaluation may indicate that a certain yield contains a systematic error, the cause of the error can be identified only rarely.

A quantitative estimate of errors by the evaluator is essential for two reasons:

	Radiochemistry		Magg	<u> </u>	Spectrome	try	
	Absolute	Relative	R-value	Spectrometry ^a	Absolute	Relative	<b>R-value</b>
Number of fissions	×			x	x	**	
Normalization to standard							
yield	-	x	x	x		x	x
Normalization to reference							
yield	-		x		-		x
Processing losses	x	x	x	x		-	
Contamination	x	x	x	x	x	x	x
Decay data	x	x	-	x	x	x	
Neutron capture data	-	-	-	х		-	
Counting corrections, $\beta$	x	x	x	-	-		-
Counting corrections, $\gamma$	x	X	x	-	x	x	x
Error assignments(%) - maximum - minimum	20 ^b 4	20 ^b 4	$10^{b}$ 3-4 ^d	l0 ^C l ^e	10 4	5 3-4 ^d	5 2-3 ^đ

## TABLE 1 - Summary of Possible Errors and Estimates of Percent Error in a Single Yield

a Isotope dilution or isobaric coupling.

- b For radiochemical data measured about 1950. Errors for intermediate dates are interpolated linearly.
- c Based on the evaluation of mass spectrometric data in [7]. Most data with errors this large were discarded.

d Minimum errors will depend on the error assigned to the normalizing value.

e The estimated error in normalizing evaluated data to 100% [7]. Mass spectrometric yields will also include in their errors the uncertainties in isotopic dilution or isobaric coupling and in isotopic abundances. First, the errors assigned by the experimenters are based on a variety of criteria. They may include an estimate of systematic errors or simply represent the statistical uncertainty in multiple measurements. The error may be the average deviation, standard deviation in the mean, the 95% confidence limit or simply half the spread between maximum and minimum results. These variations can be reduced if the evaluator assigns his own errors.

Second, a set of yields may disagree by much more than indicated by the assigned errors, indicating the presence of unidentified systematic errors. If these cannot be isolated by closer examination of the data, the most satisfactory way to deal with the data is to increase the assigned errors until the discrepancy "disappears".

Table 1 summarizes the sources of error described in the preceding sections, and then gives the percentage errors recommended for the various types of measurement. These errors are to be treated as root mean square (rms) deviations.

The procedure recommended is to assign errors to radiometric and other yields with a partial linear time dependence, say from 4% for recent measurements to 10% for early (c.1950) measurements. If these are not sufficient to account for the differences in values after rejecting obviously errant values, the upper limit should be increased so the range covers 4% to 20%. Finally, if this is still insufficient, all errors should be increased by equal amounts, added in quadrature, until the assigned errors do account for the differences.

#### 3. EVALUATION OF CHAIN YIELDS*

In determining chain yields the evaluator must deal with a very large amount of data obtained by a variety of methods. For some fissile nuclides and some masses the yields have been measured many times, for others, never. Some data agree, others differ by many times the claimed error. How should the evaluator proceed?

The solution, in essence, is to retain the special contribution of mass spectrometric data, namely, the accurately known ratios of isotopic abundances, while at the same time making full use of the radiochemical and  $\gamma$ -spectrometric yields. A simple example illustrating how a mass-by-mass analysis can affect yield ratios is given below for a hypothetical element with two fission product isotopes.

* In this section no distinction is made between chain yield and total yield at a given mass. Only for mass 136, where the shielded isobar ¹³⁶Cs has a direct yield of ~1% of the chain yield for ²³³U and ²³⁹Pu fission, will the two differ significantly.

Mass spectrometric data	Ratio of yields Sum of yields(%) Each yield(%)	1.00±0.01 10.0 ±0.5 5.00±0.255
Radiochemical yields(%)	l st isotope 2 nd isotope	4.40±0.30 5.70±0.35
Weighted mean yields(%)	l st isotope 2 nd isotope	4.74±0.19 5.24±0.21
	ratio	1.10±0.6

In this case the final ratio differs from the measured ratio by 10 times the uncertainty in the latter. The reason is that the mass-by-mass evaluation takes no direct account of the measured ratio.

On the other hand the radiochemical data contains useful information about both the relative yields and total yield which should not be ignored. In the illustration the ratio of radiochemical yields is 1.19±.11 and the sum is (10.10±0.46)%, so that the weighted means of these and the mass spectrometric values are 1.009 and 10.05% respectively, and the individual yields are 5.00% and 5.05%. These would be better values to use than either the mass-by-mass weighted means or the straight mass spectrometric data.

In real life the radiochemical data is not so complete and the solution is not so simple. How the evaluation should proceed in this case to obtain analogous results is discussed in the following sections.

## 3.1 Mass Spectrometric Yield Measurements

Mass spectrometric yields have been evaluated in two recent reports [7, 8]. Both proceeded as follows:

- (i) Corrections for β-decay and neutron capture were brought up to date if sufficient information was available. If updating was not possible the associated uncertainty in the value was estimated and the value was accepted, accepted with reduced weight or rejected on the basis of this estimate. Details of these corrections are given in [7].
- (ii) The relative abundances of the isotopes of each fission product element were determined by comparing all relevant mass spectrometric measurements. Some values were rejected because they differed from the average of the remainder for that isotope by several times the root mean square (rms) deviation from that average.
- (iii) The number of atoms of each fission product element were determined relative to a chosen standard element (e.g. neodymium) using isotope dilution and isobaric coupling measurements. Again a few values were rejected.
- (iv) The mass spectrometric yields for the light and heavy mass peaks were normalized by equating them to 100% less the sum of radiochemical and inter-

polated yields. These non mass spectrometric yields contribute from about 3% (²³⁵U heavy masses) to 20% (²³⁹Pu light masses), except for ²⁴¹Pu light masses where they contribute 55%. If this sum is accurate to 10% then the final normalization will be accurate to 2%, except for the ²⁴¹Pu light masses. A 2% uncertainty is comparable to that in normalization if the number of fissions were large enough to be measured by mass spectrometry of the fissile material. The two methods can be compared for ²³⁵U and ²³³U fission [7] and agree to 1% or better.

For most isotopic abundances of elements in the range Kr to Ru and Xe to Sm there are two or more measurements available for thermal neutron fission of ²³³U, ²³⁵U and ²³⁹Pu so that errors can be estimated from differences in measured values. These errors are less than or about equal to 1% of the value in most cases, which is consistent with the occurrence of small or negligible systematic errors in these measurements. Where only one measurement is available the error can be estimated on the basis of other results if it is assumed that any systematic uncertainty remains small.

There are fewer data available for relative element yields and the agreement is poorer. An error of two percent is typical.

#### 3.2 Non Mass Spectrometric Yield Measurements

Yields measured by radiochemical,  $\gamma$ -spectrometric and miscellaneous methods do not have the relative accuracy between masses that is a feature of mass spectrometric isotopic abundances and are best treated on a mass-by-mass basis. This can be done separately from the mass spectrometric analysis and then the two sets can be combined.

At each mass all non mass spectrometric yields should be compared, clearly discrepant values rejected and weighted means calculated using evaluator-assigned errors as discussed in section 2.3.8.

Where no mass spectrometric measurements are available this is as far as one can proceed normally, although there are a variety of less decisive tests, described in section 3.3 which may assist in choosing between two or three values at one mass when the discrepancy is large.

If both types of measurements have been made in a given mass range the greater precision of relative abundances can be brought into play as follows: the weighted mean of the non mass spectrometric yields for a given mass is divided by the isotopic abundance for that mass to obtain a set of yields corresponding to element yields as measured by mass spectrometer. The input data, non mass spectrometric yields and mass spectrometric isotopic abundances, can be adjusted to minimize the uncertainty in the "element" yield. The two element yields can then be averaged. These steps are analogous to taking weighted averages of the relative yields and the sum of the yields in the example given in section 3.

Once correctable systematic errors have been corrected and errors assigned to account for recognized systematic errors which cannot be corrected (e.g. a known change in half-life, but no statement of irradiation or decay times, or the magnitude of the original correction) it should be possible to perform the remaining steps in evaluation by computer. An example using data for yields at masses 140, 141, 142 and 144 from  235 U thermal fission is given in Table 2.

<u>Step 1:</u> The evaluated mass spectrometric data for Ce [7] are listed. The procedures followed in obtaining these (section 3.1) are also amenable to computer treatment. The isotopic abundances are averages of five sets of measurements for masses 140, 142, and 144. There is only one measurement for mass 141.

The errors quoted are the rms deviations of a single value from the mean, i.e.  $\begin{bmatrix} \Sigma^n & (x_1 - \bar{x})^2 / (n-1) \end{bmatrix}^{\frac{1}{2}}$ . Here  $x_1$  is an individual measurement,  $\bar{x}$  is the average, and n is the number of measurements. For ¹⁺¹Ce (one measurement) an uncertainty of 2% in the isotopic abundance is estimated.

The total Ce yield is based on two isotope dilution measurements plus isobaric coupling at mass 144, all three giving the ratio of Ce to Nd with an uncertainty of 1%.

The non mass spectrometric data are taken from the most recent Meek and Rider compilation [9]. Yield ratios that include ¹⁴⁰Ba are used to obtain a value for the other mass since the numerous ¹⁴⁰Ba yields agree well.^{*} A value of 6.36% for ¹⁴⁰Ba is used. Other measured yield ratios are normalized to the recommended yields of [7].

To be directly comparable, measured yields of radioactive isobars should be corrected to take account of the fact that the cumulative yields may decrease with decreasing atomic number (Z) because of significant direct yields to higher Z isobars. For most nuclides used to determine chain yields the estimated fractional cumulative yield is greater than 0.99 of the chain yield. This is the case for masses 140 - 144 in  235 U fission so no correction is needed. For the few cases where the cumulative yield is <0.99 of the chain yield the measured cumulative yield should be divided by the fractional cumulative yield to obtain a value of the chain yield.

The errors assigned by the experimenter follow the yield preceded by t. Errors assigned in this evaluation follow in brackets.

Step 2: Here we are concerned with rejection of discrepant data. Assigned errors depend too greatly on

These yields were later found to require a variety of corrections as shown in Appendix C. The agreement is not as good as indicated in Table 2.

TAR. 7	Ţ	TTAMDT.P	OF	EVALUATION	USTNO	230,,	VIRID	האתת
THOUS &	- 4	- and -	VE	PANNALION	DOTUG	<u> </u>	IICHD	DATA

Mass Chain	a 140*	a 141	a 142	a 144	Total
<pre>1. Starting Values Evaluated mass } Ce Isotopic abundances spectrometric data Ce Yields (%) Non-mass spectrometric yields - renormalized values (%) ± original error with evaluator assigned error in brackets</pre>	$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	$\begin{array}{c cccc} 7 & 0.2482 \pm 0 & 0050 \\ & (5.86 \pm 0.15) \\ \hline \\ 36 & 6.0 & (0.6) \\ 15 & 5.46 \pm 0.34(0.34) \\ 18 & 5.76 \pm 0.17(0.23) \\ 21 & 5.81 \pm 0.06(0.17) \\ 32 & 5.87 \pm 0.28(0.31) \\ \hline \\ 137 & 6.37 \pm 0.14(0.52) \\ \hline \\ 138 & 5.45 \pm 0.32(0.38) \\ 39 & 4.6 \\ \end{array}$	7 0.2522 $\pm$ 0.0043 (5 96 $\pm$ 0.13) 18 <u>La</u> 5.70 $\pm$ 0.21(0.23) <u>Ba</u> 5.08 $\pm$ 0.43(0.43)	7 0.2292+0.0030 (5.41+0.11) 36 5.30 (0.53) 18 5 79+0.43(0 43)	23.62 <u>+</u> 0.33
2. Average non-mass spectrometric yields with rms deviation (a) All values (%) (b) With omissions (%) omitted values (%)	6.26±0.20 6.32±0.08 5.77	5.67±0.52 5.82±0.32 4.6	5.39±0.44 	5.50 <u>+</u> 0.26 	
<ol> <li>Weighted mean of nms yields with omissions, using evaluator-assigned errors; <u>+</u> error based on weights; in brackets, weighted deviation from mean</li> </ol>	6.34 <u>+</u> 0.35 ( <u>+</u> 0.05)	5.78 <u>+</u> 0.26 ( <u>+</u> 0.20)	5.56+0.20 ( <u>+</u> 0.23)	5.44+0.20 (±0.13)	
<ol> <li>LSF fit using nms yields of step 3         Isotopic abundances         Total yield (%)     </li> </ol>	0.2717 <u>+</u> 0.0024	0.2481±0.0046	0,2503 <u>+</u> 0,0028	0.2299 <u>+</u> 0.0024	23.37 <u>+</u> 0.33
5. Weighted mean of nms and mass spectrometric total yields (%)	(6.39)	(5.83)	(5.88)	(5,40)	23.50 <u>+</u> 0,23
Results from mass-by-mass evaluation Crouch [6] Meek and Rider [5] Meek and Rider [9]	0.2711±0.0025 0.2679±0.0030 0.2714±0.00	0.2445±0.0075 0.2487±0.0075 0.2394±0.00	0.2518±0.0035 0.2525±0.0028 0.2549±0.00	0.2325±0.0050 0.2309±0.0026 0.2343±0.00	23.31±0.21 23.57±0.19 23.78±0.00

* A revised set of ¹⁺⁰Ba input data is given in Appendix C. Values in steps 4 and 5 have been corrected to agree with the revised mass 140 chain yield ((6.39 ± 0.10)%). (a) References for listed yields.

personal judgment to be reliable for this purpose so the data are simply averaged and the rms deviation calculated. The most deviant result is then omitted and the process repeated. If the omitted value differs from the average by more than three times the rms deviation it is not included in any of the following steps.

There seems to be no logical case for retaining such a discrepant value. Either it is wrong, due to a systematic error, the other measurements of the same yield are wrong, or none of them is right. In any case, no mixture obtained by judicious weighting is likely to give the correct answer. If rejections were done by computer the omitted results would be noted and the evaluator would make a personal judgment on the validity of the rejection before accepting the results.

In Table 2, two values are deleted by this criterion. At masses 142, with only 2 values available, the rejection criterion is inoperative.

Step 3: Weights are taken as the inverse squares of the evaluator-assigned errors and the weighted means calculated. Two errors are calculated for each mean, both approximately equivalent to the rms deviation. The first, following the t sign, is based on the weights, w, and is given by

$$\mathbf{e}_{w} = \left[ (n-1) / \sum_{i=1}^{n} w_{i} \right] / 2$$

The second, enclosed in brackets, is based on deviations of the individual values from  $\bar{x}_w$ , the weighted mean. It is given by

$$e_{d} = \begin{bmatrix} \sum w_{i} (x_{i} - \overline{x}_{w})^{2} / \sum w_{i} \\ i \end{bmatrix}^{\frac{1}{2}}$$

If  $e_d$  is significantly greater than  $e_w$ , say 30%, the assigned errors should be increased following the pattern recommended in section 2.3.8 until  $e_d \approx e_w$ .

If  $e_W^{>>}e_d$ , as for ¹⁴⁰Ba, the reverse procedure could be followed, but this is unlikely to change the weighted mean appreciably. To make  $e_W^{\approx}e_d$  in the case of ¹⁴⁰Ba, the errors assigned each value would be about 1%.

In subsequent steps the weighted means carry the larger error except for  140 Ba, where it is set at ±0.10.

Step 4: The weighted means of the non mass spectrometric yields from step 3 can now be combined with the mass spectrometric data. Dividing the weighted non mass spectrometric yields by the isotopic abundances from step 1 gives four estimates of the total yield of the four masses. These are 23.44±0.36%, 23.29±1.15%, 22.05±0.87% and 23.73±0.93% for masses 140 to 144 respectively. These values could be combined with the mass spectrometric total yield to obtain a best value, but this would not take account of differences between ratios of the weighted means yields of step 3 and the isotopic abundances measured mass spectrometrically.

To do this both sets of data with their errors, are supplied to an iterative least squares program, LSF[40], which adjusts them to minimize the error in the non mass spectrometric total yield. The results are shown. The isotopic abundances have changed significantly to allow for the non mass spectrometric yield ratios, but by less than their assigned errors.

Step 5: The weighted mean of the two total yields (step 1 and 4) is calculated. The values in brackets on the same line are the chain yields for the four masses. They are the product of the total yield, 23.50 and the isotopic abundances of step 4.

The table is completed with 3 sets of evaluated data. Of these only one [9] is based on the complete set of non mass spectrometric data given in Table 2. Since the other two include all the mass spectrometric yields these predominate in the least squares fit, especially for masses 141, 142 and 144, so that their results would not be expected to differ greatly from the input mass spectrometric data of step 1.

## 3.3 Completing the Evaluation

## 3.3.1 Relative Yields and Iterations

Many non mass spectrometric yields are relative, and must be normalized to a preliminary value for the standard yield for inclusion in step 1 of Table 2. The final value of the standard yield may differ from the preliminary value. If this difference exceeds a pre-selected limit, say 0.5%, the relative yields should be recalculated and the evaluation repeated.

Relative yields may serve another purpose. If a systematic error occurred in the preparation or counting of the standard in one measurement of relative yields, or if the final value for the standard yield is influenced by systematic errors in one or more of the measurements on which it is based, calculating the ratios of each value derived from a relative yield to the recommended value for that yield may indicate the presence of the error.

For example, if the average yield ratio for all relative yields in one set of measurements differs significantly from unity the first type of error is indicated. If the average yield ratio for all relative yields based on one standard yield differs significantly from unity the second type of error may be the cause.

In both cases the evaluator should re-examine the data. The simplest solution for the first type of error is to use another nuclide as standard. Yields having the second type of error will be difficult to locate since they would have been eliminated earlier in the evaluation if clearly discrepant.

## 3.3.2 Estimated Chain Yields

After completing the evaluation of all measured yields the most vital remaining requirement is to estimate the chain yields for masses where there are no measurements. The theoretical and empirical methods available for making these estimates are discussed by Musgrove et al. [4].

#### 3.3.3 Normalization

The chain yields for the light and heavy fragments should each add to 100%, i.e. 2 fragments per fission. The correct division between light and heavy masses will depend on  $\bar{\nu}$ , the average number of prompt neutrons emitted by fragments hear symmetric fission, i.e.

light masses <  $(A_f - \bar{v}_{sym})/2$  < heavy masses

where  $A_f$  is the mass of the fissioning (compound) nucleus.

Experimental results for neutron emission are uncertain in this region because of the low chain yields, but most lie between 3 and 4 neutrons per fission, at least for  $^{235}U$ fission [10]. A value of  $\bar{v}_{sym}$ =3.5 is suggested for normalization.

#### 3.3.4 Yield Symmetry

If all fragments emitted the same number of neutrons, the curves fitted to the light and heavy mass yields would superpose almost perfectly on reflection about the mean fission product mass.

Significant differences observed between the reflected curves are attributed to changes in  $v_{\rm m}$ , the average number of neutrons emitted at mass m.* Differences are most apparent near the yield maxima where the yields are known most accurately, ranging up to 25% (compare yields at masses 135 and 102 in ²³⁹Pu fission). It would be surprising if much larger differences occurred in other mass ranges.

In cases where two or three yields differ by much more than the sum of their errors a comparison with the curve through the complementary mass may indicate that one value is much more likely to be incorrect. On this basis the relative mass spectrometric abundances of tin isotopes [43] were omitted in one evaluation [7].

^{*} Values of  $\overline{v}_{m}$  can be determined from a comparison of fission fragment and chain yields as described by Terrell [42]. These values of  $\overline{v}_{m}$  are in satisfactory agreement with measured neutron emissions for most masses in the case of ²³⁵U thermal fission [10].

3.3.5 Average Number of Neutrons per Fission  $(\overline{v})$ 

The average number of neutrons per fission is equal to the difference in the number of nucleons before and after fission, i.e.

$$\vec{v} = A_{\text{fission}} - \sum_{i}^{\Sigma} Y_{i}A_{i}$$

where the A's are nuclear mass numbers.

If the evaluated set of yields is correct  $\bar{v}$  calculated in this way will equal the recommended value [44]. Unfortunately the reverse is not true, nor can a significant inequality indicate in what way the yields are incorrect.

The calculated value of  $\bar{\nu}$  depends on the division into light and heavy yields discussed in section 3.3.2. The transfer of one yield from the heavy to the light group increases the calculated value by an amount comparable to the uncertainty in the recommended  $\bar{\nu}$  values.

#### 3.4 A Comparison of Recent Evaluations

This comparison is restricted to 5 recent evaluations. Lammer and Eder [8] give a good summary of earlier work. In the following section the methods used in the evaluations are discussed in the context of the proposed procedures of section 3.1 - 3.3 and the mass-by-mass comparison appears in Appendix A.

#### 3.4.1 Meek and Rider [5]

This is the 5th revision of the earliest available computerized data library and evaluation. The data library is intended to include all yield measurements* and some evaluations. It appears to have achieved this aim. The data is not yet free of errors since it has accumulated a number of duplications and outdated values. Many of these were eliminated in succeeding revisions and the process is still going on.

Only a few corrections for changes in half-life and cross section are included in this edition. Each yield in the data file has both a measurer-assigned and evaluator-assigned error. Lower limits for the latter are 1% for mass spectrometric yields, 4% for other measured yields, and 10% for estimated yields. Yields considered discrepant are rejected. If there are no measured yields, an estimate is inserted as input data.

For each mass the weighted mean is calculated using as weights the inverse squares of the evaluator-assigned errors. All measured yields and most evaluated yields are included. The influence of the latter is usually minimized by assigning large errors.

^{*} For fission by thermal, reactor and high energy neutrons; direct, cumulative and chain.

Mass spectrometric relative abundances are not treated separately as recommended in section 3.2. However relative yields, in which they are included, are normalized as discussed in section 3.3.1 and the evaluation is iterative.

After each iteration the light and heavy yields are normalized to 100%. The separation between the two groups is at  $(A_{r}-\bar{\nu})/2$ . The average difference from 100% for the four thermally fissile nuclides before normalization is 0.7%, the greatest being +2.0% for the ²³³U light masses.

Relative yield measurements influence the standard yields through the use of inverse ratios. These are treated as regular input data rather than as described in section 3.3.1. The inverse ratios are generally assigned large errors, apparently to avoid divergence or oscillation in successive iterations. For this reason they do not have much influence on the final values of the standard yields.

#### 3.4.2 Crouch [6]

A computer-based data library and interrogation program have been set up under the auspices of the U.K. Chemical Nuclear Data Committee [45, 46]. A computerized evaluation procedure has been described [48] but is not yet in operation.

The data library is intended to include only measured yields that are reasonably well documented. If two results by the same author appear to be derived from the same experimental data the earlier one is omitted. No corrections for changes in half-life or cross sections are mentioned. Relative yields are normalized to preliminary estimates of the standard yields.

Errors are evaluator-assigned. Rejected values are not indicated. A computer program is used to obtain the simple average with its standard deviation and a weighted average with two errors, the one derived from the assigned weights, the other from the weighted mean deviation. Weights are the inverse squares of the assigned errors. The recommended value is usually the weighted mean and the assigned error is the greatest of the three above.

The sum of recommended values is not normalized to 100% for the light and heavy mass peaks. The average absolute difference is 1.3% and the largest is -3.35% for the ²³⁹Pu light masses.

The compilation still shows effects that can be attributed to the comparatively brief period the work has been under way. Incorrect yield values and dubious error assignments are noted in Appendix A where they affect the recommended value significantly.

## 3.4.3 Walker [7]

The methods used are an improvement of those described at Helsinki [13] for ²³⁵U fission, with the work extended to include ²³³U, ²³⁹Pu and ²⁴¹Pu. These evaluations are based on an assessment of the mass spectrometric data as described in section 3.1. Fewer radiometric yields are listed than are available in Meek and Rider [9]. They are used primarily to establish yields where there are no mass spectrometric measurements. The remaining yields, for which there are no measured values, are obtained by fitting a curve to the measured data using reflected complementary yields as a guide wherever possible.

The sum of the light and heavy mass yields, divided at  $(A_{f}-v)/2$ , are made approximately equal to 100% by renormalizing the mass spectrometric yields. The average difference from 100% before final normalization is 0.67%, and the largest is 1.2% for the ²³⁵U heavy masses.

#### 3.4.4 Lammer and Eder [8]

This evaluation of fission product yields began at Seibersdorf in 1969 as part of the research work on fuel burnup analysis. Yields from thermal neutron fission of  241 Pu are not included.

The evaluation is based on mass spectrometric measurements, as in the preceding case [7], and the methods used are very similar. The description of section 3.1 applies. Here also non mass spectrometric yields are used primarily to obtain values where there is no mass spectrometric data.

In their initial evaluation the sum of yields in the light and heavy mass peaks differed from 100% by 1 or 2%. In renormalizing they retain the mass spectrometrically measured relative light and heavy element ratios and normalize to give light mass yields that sum to 100%. For ²³³U yields additional adjustments were made in interpolated yields to make the heavy mass yield 100% as well.

#### 3.4.5 Meek and Rider (ENDF/B(IV)) [9]

In 1972, the Cross Section Evaluation Group, which is responsible for selecting data to be used in the Evaluated Nuclear Data File (ENDF/B), appointed a task force to prepare, for ENDF/B(IV), a set of fission product data to be used in burnup and heating calculations.

A committee of the task force dealing with cumulative yields* decided that for future evaluations the computerized approach of Meek and Rider would be most satisfactory provided agreed methods of error assignment and evaluation could be developed. In the short term, for ENDF/B(IV), the following changes were made in the Meek and Rider data to eliminate the main differences between their interim evaluation and the evaluation in reference[7]:

(1) The mass spectrometric data were corrected for neutron capture and  $\beta$ -decay as recommended by Walker [7]. Most of the yields not used in [7] were also omitted in this evaluation.

^{*} Members participating were T.R. England (Los Alamos), R.P. Larsen (Argonne), W.J. Maeck (Idaho Falls), W.M. McElroy (Hanford), B.F. Rider (AE Vallecitos), and W.H. Walker (Chalk River).

- (2) Where significant differences in isotopic abundances occurred, the data were reviewed. Values responsible for the differences were assigned larger errors or not used.
- (3) The errors were changed to comply with the assignments recommended by the committee. These are similar to those of section 2.3.8.
- (4) Evaluated values were not used except where no measurements were available.** Here the estimated values of [7] were used.

All the work involved in making the necessary changes to input data and evaluation program has been done by Meek and Rider. At this writing yields from the penultimate evaluation were not available, but they are tabulated and discussed in Appendix A. The final result, unfortunately, will not be ready until January, 1974.

## 3.5 Uncertainties in Current Data Requirements for Additional Measurements

In deciding what additional measurements are needed two questions should be considered -

For what masses are the yields lacking, discrepant or uncertain because only a single measurement has been made?

Is the associated uncertainty in calculations significant?

The second point will depend, of course, on the type of calculation and is considered in papers 2 through 9 at this meeting.

The main sources of uncertainty have been discussed to a greater or lesser extent in the evaluations of the preceding section, and in a discussion of the effects of yield uncertainties on fission product absorption calculations [17].

In the following sections a more detailed survey of uncertain data is presented and the measurements necessary to reduce the uncertainties are discussed.

## 3.5.1 Classifying Uncertain Data

Potentially the most important uncertainties are at masses for which there are no measurements but in practice there are very few unmeasured yields greater than 1% except for  241 Pu thermal fission.

^{**} This change has not been applied completely. For some small yields in the valley an evaluated value was inserted because the evaluation did not converge on the measured cumulative yield, apparently because a fractional cumulative yield was also available. Estimates have also been retained elsewhere, not necessarily to good effect. See, for example, Appendix A, note 4, for the ²³⁵U heavy masses.

In this survey mass spectrometric yields are given precedence because they cover most of the light and heavy mass peaks, and because there are more measurements available. For convenience the data presented in [7] are used. Only yields not listed there will be referenced.

Uncertain data are listed in Table 3 using the following categories*:

- (1) Nuclides for which the m.s. isotopic abundance is based on only one measurement These are usually radioactive and can be checked against the m.s. yields of their stable isobars as well as n.m.s. yields. Where ⁸⁹Y is listed it has been normalized to ⁹¹Zr using the measured ⁸⁹Y/⁹¹Y abundance ratio.
- (2) Nuclides for which the error in the average m.s. isotopic abundance exceeds 2% Since most errors in the average are ~1% or less, a significant systematic error is indicated if yields differ by appreciably more. Recommended yields are compared to n.m.s. yields where available.
- (3) Elements for which the m.s. element yield is based on one measurement
- (4) Elements for which the error in the average m.s. element yield exceeds 3% For both (3) and (4) the yields of isotopes of the elements can be checked against n.m.s. yields.
- (5) Chain yields based on n.m.s. measurements only, the yield exceeding 0.5% Smaller yields may be poorly measured but the uncertainty is not likely to be important in calculations. If more than one measurement has been made the highest and lowest yield values are shown.
- (6) Masses for which no yields have been measured These are listed with their estimated yields to indicate their importance. The total of estimated yields is also listed.

## 3.5.2 ²³⁵U Yields

The uncertainty likely to cause most trouble is that for the Sm yield since two isotopes (149 and 151) have very large cross sections. Although the ¹⁴⁷Sm yield agrees with that for ¹⁴⁷Nd the latter is the result of a single m.s. measurement. Radiometric yields at masses 147 and 149 also agree, but their accuracy is not high. Additional isotope dilution determinations of the element yield are required as well as another determination of the ¹⁴⁷Nd relative abundance and a  $\gamma$ -spectrometric measurement of the mass 149 yield.

The yield of 103R , an important non-saturating fission

* Here and in Table 3, m.s. = mass spectrometric, n.m.s. = non mass spectrometric. product which was previously assigned an error of 6 1/2% [7], has recently been measured by  $\gamma$ -spectrometry [18] [21] [35]. The average value is 3.09, with an r.m.s. error of 4.4% (¹⁰⁵Ru) Additional measurements are still required.

Yields at masses 129 and 130 show a large spread and should be remeasured. In addition, as many as possible of yields in groups [6] should be measured.

## 3.5.3 ²³³U Yields

The number of yield measurements for ²³³U is much smaller than ²³⁵U. No major discrepancies are apparent. The disagreements in mass spectrometric measurement for ¹⁰²Ru, ¹⁰⁴Ru and ¹⁵⁰Nd may be due to contamination but apparently not by the naturally occurring element [7]. The accuracy of the ¹⁴⁸Nd yield, of possible importance in burnup measurements, does not seem to be affected.

The low  $\gamma$ -spectrometric yield for ¹³⁵Xe [19] should be weighed against three measurements of the ¹³⁵I yield which agree with the value obtained by deducting the large direct Xe yield (see section 4.4) from the mass spectrometric yield. It is possible that the  $\gamma$ -spectrometric measurement was affected by failing to take this direct yield into account. Because of the importance of ¹³⁵Xe in thermal reactors, additional measurements are recommended, with either mass or gamma spectrometer.

There is a serious gap in our knowledge of yields from mass 102 to 131 inclusive. The mass 103 and 106 yields should certainly be measured again. Also the yields from mass 126 to 130 should be determined since their estimated values contribute the bulk of the total estimated yield.

## 3.5.4 ²³⁹Pu Yields

There are more measurements of ²³⁹Pu yields than of ²³³U yields but the agreement is much poorer. This may be because of unexpected difficulties in Pu chemistry, particularly in the earlier radiometric measurements.

The mass spectrometric yields of Ba should be remeasured. All three experimenters reported large corrections for natural Ba contamination, identified by the presence of ¹³⁶Ba, apparently present on the mass spectrometer filament. It does not appear possible that these corrections could have been in error enough to explain the 15% spread in measured yields. The isobaric coupling technique would avoid some difficulties, but there is also a 7% difference for the ¹³⁸Ba yield from ²⁴¹Pu where two such links were used. R-value  $\gamma$ -spectrometry using ¹³⁸Cs would give a more direct check.

There are two recent sets of  $\gamma$ -spectrometric ²³⁹Pu yields [20][35] which can be used to check some of the single m.s. yields noted in Table 3. The results of Ramaniah [20] are R-values, using ²³⁵U yields as reference. Those of Larsen et al.[35] are given as absolute ²³⁵U and ²³⁹Pu yields, but are here converted to R-values. In this way all dependence on  $\gamma$ -ray emission probabilities is eliminated. Their yields are compared below with the recommended yields of [7] which are based on mass spectrometric data only. The values shown are the ratios  $y_1[35]/y_1[7]$  with both sets of yields normalized to the same value at mass 140, and  $y_1[20]/y_1[7]$  with both sets normalized at mass 144.*

Mass	<u>95</u>	<u>97</u>	<u> 103</u>	106	<u>131</u>	132	<u>133</u>	<u>135</u>	142	<u>143</u>
[35]/[7]	0.95	1.01	1.22		1.09		-			
[20]/[7]	1.05	0.99	1.12	0.91	1.08	1.02	1.03	0.96	1.11	0.90
No. of m measure-	.s.									
ments	1	1	1	2	2	2	1	1	4	5

* Different normalizing yields are used because the mass 140 yield of [20] appears to be about 7% too low, while reference [35] has no mass 144 yield. Masses 140 and 144 are chosen because they are both measured mass spectrometrically as isotopes of Ce.

#### TABLE 3

CHAIN YIELDS BASED ON SINGLE, DISCREPANT OR NIL MEAS	UREMENTS							
(1) U ISOTOPES								
23511	2330							
a single m.s. isotopic abundance.								
89Sr 83Y 91Sr 95Zr 95Mo <u>Yes</u> Yes Yes Yes Yes <u>Yes</u> Yes Yes -111414 137Ra 139Ra 140Ra 141Ca 147Md	R9Sr         90Zr         95Zr         136Xe         135Cs            Zr         Y65							
Yes Yos Yes Yes Yes Yes Yes Yes Yes 1\$ -2\$ -1\$ -5\$ 6\$								
isotopic abundances agree poorly (assigned error >+ 2%	)							
1545m (2 7%) (one measurement rejected)	¹⁰² Ru(2 6%) ¹⁰⁴ Ru(2.7%) ¹⁰⁵ Nd(3 8%) Yes 3%							
(3) Element yields based on a single m s measurement.								
Kr         Xe           83         84         85         131         132         133           Yes         Yes         Yes         No         Yes         Yes           *5%         7%         -3%[18]         1%[18][21]         5%[18]         2%[18]	Kr         Rb         Ru         Xe           83         85         106         131         132           Yes         No         Yes         Yes         Yes           5%{50}         +14%{19}         14%         -5%         -10%							
(4) Element yields that agree poorly (assigned error >+3\$).								
Sm (61)         Fu (61)           147         149         153           Yes         Yes         Yes/No (2 values)           -61         -41 (both including [18])         -51/-131	None							
n m,s, measurements only								
99         103         105         129         130           6.244.12         3         22+.09[18]         .83+.20         1         12+.18         2.0+0.5           6.037         08         2.957         09[35]         .83+.20         S0+04[49]         2.0+0.5	$\begin{array}{cccccccccccccccccccccccccccccccccccc$							
measurements available.								
80         82         108         110         113         114         116         - 120           12         33         .07         .022         .012         .100        010	78 79 80 82 107 108 .06 .16 .26 .60 13 .07							
122 124 126 Total 013 .020 .053 0 7	110     113     114     116     122     123     124       .029     015     .014     .014     .025     .037     .060       126     128     129     130     155     Total       26     1<0							
	$\frac{\text{CHAIN YIELDS BASED ON SINCLE, DISCREPANT OR NIL MEAS}{(1) U ISOTOPES}$ $\frac{2^{35}\text{U}}{2^{35}\text{U}}$ a single m.s. isotopic abundance. $\frac{89}{785}  \frac{89}{785}  \frac{91}{785}  \frac{95}{785}  \frac{95}{785}  \frac{95}{785}  \frac{95}{785}  \frac{95}{785}  \frac{95}{785}  \frac{95}{785}  \frac{95}{785}  \frac{91}{785}  \frac{91}{78}  \frac{91}{140} \text{Ba}  \frac{11}{11} \text{Ce}  \frac{117}{7} \text{Nd}  \frac{-11}{7} \text{Nd}  \frac{-11}{7} \text{Nd}  \frac{-11}{7} \text{Ce}  \frac{117}{7} \text{Ba}  \frac{137}{78}  \frac{137}{78}  \frac{137}{78}  \frac{140}{98}  \frac{111}{126}  \frac{117}{7} \text{Nd}  \frac{-11}{7} \text{Ce}  \frac{117}{7} \text{Nd}  \frac{-11}{7} \text{Ce}  \frac{11}{7} \text{Ce}  \frac{117}{7} \text{Nd}  \frac{-11}{7} \text{Ce}  \frac{11}{7} $							

a M.s yields can be checked as follows if the nuclide is stable the only check possible is an n.m.s yield of a radioactive isobar, if the nuclide is radioactive there may be any or all of the following - an m.s. yield for the stable isobar, an n.m.s. yield of the nuclide itself or an n.m.s. yield of a radioactive isobar. "Yes" shows that the check agrees with the yield being checked, "No" that it does not. A blank (--) shows that no check is available.

b If the uncertainty is "X%", then the check, or average if there is more than one, has an uncertainty of X% and is close to the yield being checked. If the uncertainty is "+Y%" of "-Y%" then the check, or average if there is more than one, is Y% greater or less than the yield being checked If "Yes" in the line above, the uncertainty in the check exceeds the difference.

c The value recommended is 0 53%

For masses 95 and 97, measured mass spectrometrically as Mo isotopes, the  $\gamma$ -spectrometric yields support the m.s. values, although the two  $\gamma$ -spectrometric measurements differ by 10% at mass 95.

There are appreciable discrepancies in the mass spectrometric measurements of Ru. The two isotopic abundances for ¹⁰⁶Ru differ by 8 1/2% while the two measurements of element yield differ by 4%. The ¹⁰³Ru yield is based on a single measurement of the 103/106 ratio that was corrected in [7] by a 7.7% increase, tentatively ascribed to a calculational error in the original paper.

If the high isotopic abundance of ¹⁰⁶Ru and the high element yield were both omitted the mass spectrometric yield for mass 106 would decrease by more than 6% and would then be in reasonable agreement with the single  $\gamma$ -spectrometric value. However, the yield for mass 103, which is tied to mass 106 by the ratio measurement, would also decrease by

#### TABLE 3

## CHAIN YIELDS BASED ON SINGLE, DISCREPANT OR NIL MEASUREMENTS

(2) <u>Pu ISOTOPES</u>

Fissile Isotope	239 Pu	243 pu					
(1) Yields based on only a	single m.s. isotopic abundance						
Fission product Checks from other im.s. yields ^a Uncertainty in checks ^b Fission product Checks from other im.s. yields ^a Uncertainty in checks ^b	89St         89Y         95Zr         95Mo         97Mo         96Mo           differ by 54         Used to normalize         -         -         -         -         -         -         -         -         -         -         -         -         -         -         -         -         -         -         -         -         -         -         -         -         -         -         -         -         -         -         -         -         -         -         -         -         -         -         -         -         -         -         -         -         -         -         -         -         -         -         -         -         -         -         -         -         -         -         -         -         -         -         -         -         -         -         -         -         -         -         -         -         -         -         -         -         -         -         -         -         -         -         -         -         -         -         -         -         -         -         -         -         -         -         -         - </td <td>All isotopes of Kr, Rb, Sr, 2r and Ru. The few n.m.s. measts. available agree with recommended yields except for mass 91 (-101) TSSXe TSSC TSSC TSSC TSSC TSSC TSSC TSSC TSS</td>	All isotopes of Kr, Rb, Sr, 2r and Ru. The few n.m.s. measts. available agree with recommended yields except for mass 91 (-101) TSSXe TSSC TSSC TSSC TSSC TSSC TSSC TSSC TSS					
(2) Yields for which m s. i Nuclide (error) n.m.s. checka Uncertainty in check ^b	sotopic abundances agree poorly (assigned error > 22)       106Ru (4.3%)     135CS (2 2%)       No     Yes (sum of 135, 136 agree Text       No     Yes ments	¹⁵⁰ Nd (3.0¢)					
(3) Element yields based on	a single m s. measurement						
Element Isotopes with n.m.s. checks n.m.s. checks ^a Uncertainty in check ^b	Mo See (1) above Note Xe/Kr ratio does not sgree [51]	Kr, Rb, Sr, Zn, Ru <u>Sma</u> See (1) above 147 149 151 No Yes Yes +7% -4% -4%					
(4) Element vields that agree poorly (assigned error >±3%)							
Eloment	Ba Two values agree, assigned error +2 1% Third value is 15% greater. No n.m s. check at ma≤s 138	Ba Two values differ by 7.5%. Single R-value measurement at mass 138 depends on 2 ³⁹ Fu yield for 1 ³⁸ Ba (also uncertain).					
(5) Yields >0.5% based on a	n.m.s. measurements only						
Mass Highest value (%) Lowest value (%)	99         105         109         139         141           6.47+.18         5.47+.06         1.56+.20         5.98+.15         5.67+1           5.617.33         1.137.06         5.557.07         4.707.7	$\begin{array}{cccccccccccccccccccccccccccccccccccc$					
(6) Estimated yields, no measurements available							
Mass Estimated yield (%) Mass Estimated yield (%) Estimated yield (%)	$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	$ \begin{array}{cccccccccccccccccccccccccccccccccccc$					

N.s. yields can be checked as follows. if the nuclide is stable the only check possible is an n.m.s. yield of a radioactive isobar; if the nuclide is radioactive there may be any or all of the following - an m.s. yield for the stable isobar, an n.m.s. yield of the nuclide itself or an n.m.s. yield of a radioactive isobar. "Yes" shows that the check agress with the yield being checked, "No" that it does not. A blank (--) shows that no check is available.

b If the uncertainty is "X\$", then the check, or average if there is more than one, has an uncertainty of X\$ and is close to the yield being checked If the uncertainty is "+X\$" or "-Y\$" then the check, or average if there is more than one, is Y\$ greater or less than the yield being checked. If "Yes" in the line above, the uncertainty in the check exceeds the difference. this amount and would then differ by about 25% from the mean of the  $\gamma$ -spectrometric values. Clearly additional mass and  $\gamma$ -spectrometric measurements are required at mass 103 and 106.

Like ¹⁰³Rh, ¹³¹Xe is an important slowly saturating neutron absorber. As with Ru, there are disagreements in the Xe m.s. yields. Two determinations of both the Kr and Xe element yields give ratios that differ by 7 1/2%. If the higher Kr yield [15] were adopted, along with the Xe/Kr ratio based on isotope dilution with a calibrated Xe/Kr spike [51], then all Xe yields (masses 131, 132, 133 and 134) would increase 5%. This would bring the m.s. yields for ¹³¹Xe into reasonable agreement with the two  $\gamma$ -spectrometer values, retain the agreement at mass 132 and give a ¹³³Xe yield (based on one m.s. measurement) equal to the  $\gamma$ -spectrometric yield of [20].

This procedure could be extended a step further. If the ¹³³Xe yield obtained above is used to give a Cs element yield via its isobar, ¹³³Cs, this element yield agrees with the value obtained by Lisman et al. [15]. Two other measurements of the Cs yield are 3% and 5% lower. The use of this higher yield would increase the discrepancy at mass 135, but here one can speculate that the  $\gamma$ -spectrometric measurement is in error because it did not take account of the large direct yield to ¹³⁵Xe.

The preceding is, of course, highly speculative, and additional mass spectrometric measurements of the Xe/Kr yield ratio and the Cs yield, as well as  $\gamma$ -spectrometric measurements at masses 131, 133 and 135, are clearly required.

The mass spectrometric yields at masses 142 and 143 are based on several measurements that agree well. Only the final normalization to 100% is in doubt because of the uncertainties in the Xe and Cs yields. If these were increased as discussed above, the remaining yields would have to be decreased by about 2%. Since this is far short of removing the disagreement it appears probable that the  $\gamma$ -spectrometric yields are in error.

Returning to Table 3, the review of n.m.s. yields in part (5) shows that there is disagreement at all masses where there is more than one measurement. The  $\gamma$ -spectrometric results of [20] when normalized at mass 144, give a mass 99 yield in good agreement with 2 of the 3 other yields, with an average value of 6.32, so that it would be reasonable to omit the low value in Table 3. For the other masses in part (5) plus masses 107 to 130 inclusive, for most of which no data is available, additional yield measurements should be made.

## 3.5.5 ²⁴¹Pu Yields

There are many fewer measurements of ²⁴¹Pu yields so that the greater number fall in the "no measurement" or "single measurement" categories. The most important region that is undermeasured is the mass range 100 to 130 which includes the maximum of the light mass peak. There is one mass spectrometric measurement, of the Ru isotopes, but it is not normalized by isotope dilution [15]. Outside this range yields at masses 100 ( $\sim$ 6.2%) and 139 ( $\sim$ 6.3%) should also be measured.

The mass spectrometric Ba yields again disagree. One value, based on two isobaric links at masses 138 and 140 [52] differs from the isotope dilution measurement by 7% [15]. If the latter is correct then the Ba isotope yield ratios normalized to it would give yields at masses 138 and 140 that differed by 7% from Xe and Ce measurements. Further measurements are also required at these masses.

At mass 135 two pile oscillator measurements [53] [54] of the ¹³⁵I yield agree well. They are 13% greater than the value obtained by subtracting the small direct Xe yield from the m.s. yield. Again further measurements are essential.

#### 4. CUMULATIVE AND DIRECT (INDEPENDENT) FRACTIONAL YIELDS

The stable and long-lived  $\beta$ -active nuclides used to determine chain yields represent a negligible fraction of those present immediately following fission and fragment de-excitation by prompt neutron and gamma emission. Most of these are short-lived  $\beta$ -active nuclides that are important in reactor heating, radiation damage, and delayed neutron production. Fortunately for reactor physicists they are also important to scientists interested in understanding the fission process, and it is the latter who have done almost all the measurements of direct and cumulative fractional yields.

#### 4.1 Cumulative Yields, Direct Yields and β-decay

The cumulative fractional yield,  $c_{ij}$ , of the ith isobar at mass j, is related to the direct fractional yields,  $d_{ij}$ , by

$$c_{ij} = \sum_{k=i}^{\infty} d_{kj}$$

A typical direct yield distribution as a function of the atomic number, Z, is given in the following hypothetical example.

Atomic no. of isobar, Z _{ij}	2 * s	z _s -1	²₅-2	^z s ^{−3}	² s ⁻⁴	2 _s -5	z _s -6
Direct yields, d _{ii}	<.003	.023	.405	.517	.054	.0007	∿0
Cumulative yield, cij	1.000	~1.00	.977	.572	∿.055	∿.0007	
Half-life of Isobar	00	∿l day	∿l hr	∿3 min	∿20 s	∿2 s	
Total Energy (MeV/decay)	∿1	∿2	∿ <b>4</b>	∿5.5	∿7	∿8.5	∿10
	Law water and the second second second second second second second second second second second second second s	the second second second second second second second second second second second second second second second s	ليستحد ويتحق يستحد والم			the second second second second second second second second second second second second second second second se	

* stable isobar

The features of main interest for reactor applications are that the direct yield distribution-in-Z peaks sharply, typically for nuclides with half-lives in the range of a few minutes to a few hours, and that the cumulative yield is close to unity for the last two  $\beta$ -decays  $(Z_S-2 \rightarrow Z_S-1 + Z_S)$ . When the reactor has run long enough for this isobaric chain to reach equilibrium the decay rate for each isobar will be proportional to its cumulative yield,  $c_{ij}$ , and the corresponding rate of energy release will be proportional to  $c_{ij}$ times the MeV/decay for that isobar.

The average  $\beta$ -decay chain length for mass j,  $\bar{n}_{\beta\,j}$ , is given by

 $\bar{n}_{\beta j} = \sum_{i} d_{ij} (Z_{sj} - Z_{ij})$   $(\bar{n}_{\beta j} = 2.61 \text{ decays/fission}$ in the example)

The average  $\beta$ -decay chain length over all masses is given by

$$\bar{n}_{\beta} = \sum y_j \bar{n}_{\beta j}/200$$
 where  $y_j$  is the chain yield of the jth mass in percent.

A value of  $n_{\beta}$  can also be deduced by equating the atomic number before fission,  $Z_{fissile}$ , to the average atomic number of the stable fission products, i.e.

$$\bar{n}_{\beta} = \sum_{j} (y_{j} Z_{sj}/200) - Z_{fissile}$$

Using the recommended yields of Appendix A the values of  $\bar{n}_{\rm g}$  are:

Fissile nuclide ²³⁵U ²³³U ²³⁹Pu ²⁴¹Pu decays/fission 6.07 5.20 5.50 6.30

Values of  $n_{\beta j}$  will average  $n_{\beta}/2$  but considerable variation is expected depending on nuclear structure, particularly the displacement of  $Z_s$  from the atomic number of the most stable nuclide for that mass.

## 4.2 Measurements of Direct and Cumulative Yields

The methods for measuring direct and cumulative yields are similar to those described in section 2, except that, for the great majority, the nuclides studied are relatively short-lived. They can be expected to include greater systematic errors.

Direct yields are measured either by isolating the nuclide before its parent decays to an appreciable extent or by measuring its cumulative yield and that of its parent. The direct yield is then just the difference  $c_{ij} - c_{(i-1)j}$ . If  $c_{(i-1)j}$  is very small  $(Z_{i-1} << Z_s)$  it may suffice to estimate its value.

Direct and cumulative yields are measured relative to some standard yield, so that the ratio of the chain yield to the same standard must also be determined before the fractional yield can be obtained.

## 4.3 Compilations of Direct and Cumulative Yields

There are several recent compilations of direct and cumulative yields. The reviews by Wahl et al. [10] and Amiel and Feldstein [12] treat only the relatively abundant ²³⁵U measurements, while Denschlag [11] also includes ²³³U and ²³⁹Pu fission.

The data library of Meek and Rider [5] [9] includes direct and cumulative yields in addition to chain yields and they are used in their evaluation of the direct yield distribution at each mass.

From the discussion and illustration of section 4.1 it can be seen that the fractional cumulative yield is the most important factor determining isobaric contributions to  $\beta$ -decay energy release and radioactivity for a given mass chain. Isobars with  $Z_{ij}$  close to  $Z_{sj}$  have values of  $C_{ij}$  close to unity and are, therefore, subject to only small uncertainties while, if  $Z_{ij} >> Z_{sj}$ , values of  $c_{ij}$  are negligibly small. Thus the main uncertainty in reactor calculations will come from isobars between these extremes. For the purpose of this meeting a survey of direct yields is required that is confined to this region of uncertainty and is up-to-date, easily read and widely accessible. None of the current surveys are satisfactory in all these respects.

Appendix B is a compilation of direct and cumulative yields for all fission products with  $y_j \ge 1\%$  and  $d_{ij} \ge 0.05$ . The first limit eliminates yields that are so small the excluded nuclides make only minor contributions in calculations, and for which there are very few data anyway. The second limit corresponds to  $1.0 > c_{ij} > 0.05$ . The data library of Meek and Rider [9] has been used with certain deletions, and with additions from the other compilations [10] [11] [12] and unpublished measurements.

It requires only a brief look at Appendix B to see that many more direct and cumulative yield measurements are required. For ²³⁵U there are some data available for about 65% of the fission products listed, but for about half of these either there is only a single measurement or the spread between values is large.

For ²³³U and ²³⁹Pu there are so few measurements that evaluation is impossible. For ²⁴¹Pu they hardly exist and its yields are not included in Appendix B. With very few exceptions, almost all direct yields for these 3 fissile nuclides must be estimated using a semi-empirical model as discussed by Musgrove et al. [41].

#### 4.4 An Evaluation for Mass 135

There are insufficient data for most fission products to carry out a satisfactory evaluation, even for  $2^{35}U$ thermal fission, and such an evaluation will not be attempted. One region where measurements are relatively abundant is between masses 131 and 135, due to the relative ease of extraction of Xe and I isotopes.

Fissile Nuclide	235U		U 8 8 2		239pu		241 <b>P</b> U	
	Cum.	Direct	Cum.	Direct	Cum.	Direct	Cum.	Direct
Cs (chain yield) Xe (Z = 54)	6.60 <u>+</u> .16 [7] .89+.09 [18]	.025+.025 [56]	6.21+.15[7] .92+.10 [19]	.099*.013*[60]	7.69+.26[7] .955+.025 [20]	.129±.022 [59]	7.06±.24 [7]	.049+.038[59]
, ,	1.02±.02[18]	.041+.012 [55]	••••••••	.154+.014 [59]		.151+.004 [61]		.0324.002[61]
		.027±.010 [57]		.218±.005 [61]				
		.035+.010 [58]						
		.024+.017 [59]						
		.019+.013 [60]						
Weishted mean	1.01+.04	.035+.0035	.92+.10	.21+.02	.955+.025	.150+.004		.032+.002
Recommended	1.00	.035	-0.99	. 21	-0.99	.150	1.00	.032
I (Z = 53)	.88+.09 [62]	.70+.17*[64]	.78+.02 [53]	.606+.024 [71]	.75+.08 [71]	.608+.017 [71]	1.05+.10 [73]	
,	.994.08 [63]	.434.10 [65]	.76+.08 [70]	1	.85+.02 [53]		1.02+.03 [53]	
	.964.07 [18]	.55+.09 [66]	.85+.07 [54]		.86+.02 [54]		1.14+.08 [54]	
		.47+.02 [67]			.25+.04+[69]		ł	
		.44+.07 [68]						
		.27+.03+[69]						
Åverage	.94+.06	.47±.05	.80 <u>+</u> .05	.606±.024	.82±.06	.608+.017	1.074.06	
Recommended	.965	.47	. 79	.61	.85	.61	.968	(45)
Te (2 = 52)	.524.07 [68]	.23±.15 [64]				.48+.04 (69)		
Ĭ	.50+.04 [74]	.50+.04 [69]						
Average	.51+.01	.37				.48+.04		
Recommended	.50	(47)	.18	(18)	.24	(23)		(48)
Sb (Z = 51)		.20 [69]				.24 [69]		
Recommended		(03)		(-0)		(01)		(04)

YIELDS OF MASS 135 ISOBARS

* Value not used in obtaining weighted mean or average (using rejection criterion of section 3.2). Sum of Xe and I. Value not used as * above. +-

TABLE 4

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Mass 135 is of particular interest because of the importance of  $^{135}Xe$  as a neutron absorber, and, in particular, the dependence of reactor stability on the direct yield of  $^{135}Xe$  and the cumulative yield of  $^{135}I$ .

The measurements are presented in Table 4 as fractional yields. Absolute yields have been converted to fractional yields by dividing by the Cs (chain) yields listed in the first line. Errors are evaluator-assigned.

The evaluation proceeds as in section 3.2 except that, for I and Te, averages are calculated rather than weighted means. The recommended cumulative yields for I are equal to the difference between the cumulative and direct yield of Xe ( $c_{Xe,135} - d_{Xe,135}$ ) since this has a smaller error than the average of the measured yields. The recommended cumulative yields for Te are obtained in the same way. In all cases the value obtained as a difference between cumulative and direct yields differs from the average of the measured cumulative yields by appreciably less than the rms deviation in the latter.

Bracketed yields are estimates extrapolated from the recommended yields of the higher 2 isobars assuming that the distribution is sharply peaked as in the example of section 4.1.

Measured yields are rejected if they differ from the average of the remainder by more than 3 times the rms deviation from the average. All rejected yields except one were obtained from the recent paper by Gunther et al. [69]. These measurements were done with an on-line double-focussing mass separator as described in section 2.2.6.

A major finding of this investigation is that the direct yield distribution-in-Z for ²³⁹Pu is almost identical to that for ²³⁵U for all masses investigated (131 to 139 inclusive). At mass 135 this is clearly in disagreement with the radiochemical and  $\gamma$ -spectrometric measurements and the pile oscillator results (section 2.2.6) for the cumulative I yields.

This can be seen more readily in Fig. 1 in which the recommended yields of Table 4 and the measurements of [69] are plotted. In the latter the photographic plates were examined before an appreciable fraction of 6.6-hr  135 I and 9.2-hr  135 Xe could decay, so that the direct yield corresponding to no observed  $\beta$ -decays includes both isobars. This is indicated in Fig. 1 by the horizontal bar.

On the basis of the mass 135 evaluation all results of reference [69] must be suspect. It is possible that the direct yields observed are dictated by the conditions of the experiment rather than the actual yields. In discussing a similar discrepancy at mass 132, Naeumann et al. [74] suggested that selection of a restricted range of fission fragment energies for analysis in the mass separator may account for the difference.



## 5. YIELDS FROM FISSION BY EPITHERMAL NEUTRONS

It is now well established that yields in the valley near symmetric fission vary appreciably with neutron energies in the resolved resonance region [75] [76] [77]. An effect can also be seen using epithermal neutrons obtained by filtering a reactor flux with Cd or Sm.

The effect can be explained empirically by the twomode fission hypothesis origninated by Turkevich and Niday in 1951 [78]. It supposes that the observed yield is composed of two distributions, an asymmetric two-humped mode which is independent of neutron energy and a symmetric mode with a maximum for symmetric fission whose magnitude varies with neutron energy. In the epithermal region this dependence on energy varies from resonance to resonance between limits which appear to be related to the fission width and, to some extent, the spin of the compound nucleus [76] [77]. At much higher neutron energies, beyond the scope of this paper, the symmetric mode increases monotonically with energy [79].

## 5.1 The Symmetric Mode in the Epithermal Fission of ²³⁹Pu

 239 Pu shows the largest yield changes attributable to resonance absorption. Cowan et al. [76] used  115 Cd and  99 Mo to monitor these changes for resonances from 15 to 82 eV. Their results fall into three groups, based on the ratio of thermal to resonance yield, with averages of 2.7t0.2, 1.8t0.1 and 0.67t0.02 for this ratio. In thermal fission about 48% of the fissions are due to the resonance at 0.3 eV and the remainder to a bound level. These contributions are compatible with a ratio of 2.7 for the 0.3 eV level and 0.67 for the bound level.

Sm, with a large resonance at 0.098 eV, can be used to filter a reactor spectrum so that most  239 Pu fissions are caused by resonances at energies from 0.3 eV up. The work of Regier et al. [80] who obtained a thermal/epi-Sm ratio of 2.41±0.15 for  115 Cd confirms this expectation.

A survey of measured thermal/epi-Sm ratios as a function of fission fragment mass will, therefore, give a good indication of the width and magnitude of the symmetric mode and the total yield of fission products that change with neutron energy. The summary by Tong et al. [81] is used.

F.P.Mass	Th/Epi-Sm Ratio	Chain Yield(%)	Epi-Sm Yield(%)	Symmetric Mode Yield(%)
72	1.44±.05[82]	0.00011[9]	0.000076	0.000034
77	1.08±.04[81]	0.0059[9]	0.00546	0.00044
112	1.32±.08[82]	0.11[7]	0.083	0.027
115	2.41±.15[80]	0.038[7]	0.016	0.022
121	2.34±.06[80]	0.038[7]	0.016	0.022
125	1.79±.06[80]	0.10[7]	0.056	0.044
166	1.45±.07[81]	0.000068[9]	0.000047	0.000021

In the table the epi-Sm yield is the quotient of the chain yield and the measured ratio, and the difference from the chain yield, shown in the last column, is attributed to the energy dependent mode. If the result for mass 125 is correct the energy dependent mode does not peak at the mean mass.

To make a rough estimate of the fraction of the total yield that varies with energy assume that the energy dependent mode is symmetric, with a constant value of 0.025% from 100 to 138, and a value that decreases linearly outside this range to the measured values at masses 72 to 166. For this distribution the fraction of the yield in the energy-dependent symmetric mode is 1.65% out of 200%, or about 0.01.

For a mass having a thermal neutron chain yield of about 6% which is typical of the yields in the light and heavy mass peaks, the epithermal yield will be about  $(6.00 - 0.025)\% \times 1.01 = 6.03\%$ . The renormalization factor, 1.01, is required because the symmetric mode decreases for epithermal fission and the yields must total 200%.

## 5.2 Peak Yields in Epithermal Fission of 239Pu

In the preceding section it was assumed that only the symmetric mode changed with neutron energy. It is also possible that peak yields will change relative to each other, contrary to the assumption of the two-mode hypothesis.

The effect of neutron energy on peak yields has recently been measured by Tong et al.[81] for  239 Pu. They measured the thermal/epi-Sm and thermal/epi-Cd ratios for a total of 19 nuclides representing 7 mass chains between masses 85 and 105 and 7 between 131 and 147 and found them to be constant with an uncertainty of less than 3% for a series of 5 irradiations (3 epi-Sm, 2 epi-Cd).*

The recent measurements of Popa et al. [83] using  $^{235}U$ under a Cd filter show significant increases in peak yields relative to thermal neutron fission. For example, they report an epi-Cd/thermal yield of 1.2 at mass 131. Their total observed increase in the heavy mass peak alone is 2.3% over thermal fission for the 9 mass chains for which results are listed. This is about four times greater than the change that could be accounted for by assuming complete loss of a symmetric mode with a peak equal to the yield at mass 117 (00.01%), i.e. by assuming that, in epi-Cd fission, valley yields are about zero.

Thus the only way in which these results could be consistent with a total yield of 200% would be for other peak yields to decrease by similar amounts. Clearly such decreases would have to be measured before the results of Popa et al.[83] can be accepted. The results of

Their results, converted to ratios relative to the mean value of the thermal/epithermal ratio for each irradiation, are given in Appendix C.
Balcarczyk et al. [84] for  ${}^{235}U$  and  ${}^{233}U$  under Cd show no significant changes at masses 137, 103 or 106, which were not measured by Popa et al. [83].

### 5.3 Effects of Epithermal Yield Variations on Fission Product Absorption

From the preceding two sections it is apparent that changes in yields due to variations in the symmetric mode are small compared to uncertainties in measurements of the ratio of thermal to epi-Sm or epi-Cd yields. This will also be true for ²³³U and ²³⁵U since changes in the peakto-valley ratio with neutron energy for those nuclides are several times smaller than for ²³⁹Pu.

To decide whether there is any significant effect on fission product absorption due to changes in yields with neutron energy, it would be necessary to measure very accurately the changes for such important absorbers as ¹⁰³Rh, ¹³¹Xe, ¹³⁵Xe, ¹⁴³Nd, ¹⁴⁷Pm, ¹⁴⁹Sm and ¹⁵¹Sm.

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### APPENDIX A

# COMPARISON OF EVALUATED YIELDS FROM THE THERMAL NEUTRON FISSION OF ²³⁵U, ²³³U, ²³⁹Pu and ²⁴¹Pu

The evaluated yields of Meek and Rider [5] [9], Crouch [6], Lammer and Eder [8] and Walker [7] are listed in Tables Al to A8. Light and heavy mass yields are tabulated separately, with light masses defined as those less than 1/2 (fissioning mass - 3.5).

The first three  $({}^{2+1}Pu)$  or four yield columns list published evaluated values with their error assignments [5] [6] [7] [8]. The next column, headed ENDF/B, is the unpublished interim set based on the Meek and Rider yield library and evaluated by them [9], as discussed in section 3.4.5.

The last yield column gives a set of recommended values. The recommended yields are averages of the preceding columns with the following exceptions:

- (i) The Meek and Rider [5] values are not included since the ENDF/B set is based on the same data library except for recent additions and corrections.
- (ii) The Crouch [6] values are used infrequently for two main reasons: the Idaho Falls values are taken from a preliminary publication [85] and often differ significantly from the final yields [15]; the sums of light and heavy mass yields do not add to 100% and will introduce a bias into the evaluation. Where these differences are particularly large a note is appended.
- (iii) Where an explanatory note is indicated in the last column.

For each set values of  $\sum_{i} y_i$ ,  $\sum_{i} y_i A_i$ ,  $\overline{A}_L$ , and  $\overline{A}_H$  are included. Here A is the nucleon number and the average values are given by  $\overline{A} = \sum_{i} y_i A_i / \sum_{i} y_i$ . The calculated value of  $\overline{\nu}$  (= fissioning mass -  $\overline{A}_L$  -  $\overline{A}_H$ ) is given at the bottom of each heavy mass table (A2, A4, A6, A8).

The recommended yields do not sum to 100%, the difference from 100% depending on how closely they follow the evaluated yields. Before use, each value should be divided by the appropriate value of  $\Sigma y_i$ .

In view of the many discrepancies discussed in section 3.5, it may seem surprising that the evaluations disagree so infrequently. This only indicates that the evaluators have usually responded in similar ways when faced with a discrepancy, and should not be taken as an indication that the problem has been resolved and the correct value determined.

Hass	Meek & Rider	Crouch	Lammer & Eder	Walker	ENDF/B	Recommended	Note
<73	0 000038 (168)	VD.000025 (15%)	~0.000025		0 000038 (16%)	0.00003	
73	0.00010 (328)	0.00010 (15%)	0.00010	Î	0.00010 (23%)	0.00010 (20%)	
74	0.00034 (328)	0.00034 (15%)	0.00035	0.005*	0.00035 (32%)	.00035 (208)	
75	0.00121 (32%)		0.001*	1	0.00125 (32%)	0.0012*	
76	0.0035 (64%)	······································	0.003*		0.0038 (458)	0.0035*	
77	0.0092 (.0007)	0.0081 (.0009)	0.008	0.0083 (.0008)	0.0077 (.0010)	0.0082 (.001)	1
78	0.0203 (.0032)	0.020 (.002)	0.020	0.020 (.002)	0.0184 (.0041)	0.020 (.002)	-
79	0 055 (.009)	0.055 (.006)	0.056	0.056 (.006)	0.058 (.001)	0.056 (.006)	
80	0.095 (.032)	0.11*	0.11*	0.120 (.024)*	0.128 (.031)	0.12*	
81	0.197 (.016)	0.21 (.02)	0.22	0.20 (.02)	0.216 (.034)	0.21 (.02)	
82	0,243 (.076)	0.333*	0.35	0.33 (.06)*	0,344 (.076)	0.34*	
83	0.535 (.005)	0.515 (.005)	0.532	0.535 (.013)	0.532 (.003)	0.533 (.013)	2
84	0.997 (.010)	0.959 (.007)	1.000	0.986 (.023)	0.987 (.010)	0.986 (.023)	2
85	1.331 (.027)	1.30 (.04)	1.328	1.33 (.03)	1.315 (.010)	1,32 (.03)	2
86	1.937 (.019)	1.89 (.13)	1.97	1.96 (.05)	1.953 (.020)	1.95 (.05)	2
87	2.553 (.051)	2.64 (.13)	2.56	2 53 (.06)	2.548 (.071)	2.55 (.07)	3
88	3.646 (.072)	3.69 (.18)	3.62	3.59 (.07)	3.640 (.072)	3.62 (.07)	3
89	4.809 (.096)	4,77 (.07)	4.84	4,74 (.09)	4.891 (.192)	4.80 (.10)	
90	5,930 (,119)	5.89 (.65)	2.91	5.82 (.11)	5.896 (.165)	5,89 (.11)	
91	5.912 (.118)	5.90 (.12)	5.93	5 95 (.11)	5.928 (.083)	5.93 (.11)	
92	5.987 (.120)	5.95 (.06)	5,98	5.98 (.07)	5.9/4 (.085)	5.97 (.07)	
93	6.407 (.128)	0.34 (.13)	6.35	6.41 (.07)	6.366 (.090)	6.40 (.07)	
94	6.449 (.129)	C AS ( 13)	6.54	6.45 (.07)	6 432 (.090)	6.44 (.0/)	
35	6 270 ( 125)	6 23 ( 12)	6 20	6 30 ( 02)	6 264 ( 088)	6 28 ( 07)	
90	5 941 ( 119)	5.87 (.15)	6.00	6.07 (.10)	5.901 (.059)	6 03 (.10)	4
98	5 788 ( 116)	5.77(.12)	5 81	5.81 (.10)	5.789 (.161)	5.79 (.10)	4
99	6 130 ( 061)	6.14 (.05)	6.11	6.14 (.09)	6,128 (.061)	6.13 (.06)	•
100	6.283 (.125)	6.24 (.19)	6.32	6.31 (.11)	6.264 (.175)	6.30 (.11)	4
101	5.097 (.102)	5.05 (.25)	5.05	5.07 (.07)	5.051 (.141)	5.05 (.07)	
102	4 206 (.084)	4.19 (.04)	4.19	4.19 (.06)	4.215 (.117)	4.19 (.06)	
103	3.103 (.124)	3.03 (.18)	2.95	3.05 (.20)	3.124 (.043)	3.12 (.04)	5
104	1.832 (.036)	1.82 (.05)	1,83	1.83 (.03)	1.826 (.051)	1.83 (.03)	_
105	0.946 (.040)	0.96 (.04)	0.90	0.95 (.20)	0.927 (.037)	0.927 (.040)	6
106	0.391 (.008)	0.39 (.05)	0,387	0.390 (.006)	0.393 (.011)	0,390 (.006)	
107	0.191 (.061)	0.166*	6.17*	0.16 (.04)	0.206 (.030)	0.17*	
108	0.0704 (.022)	0.070*	0.057*	0.070 (.014)*	0.0743 (.022)	0.070*	
109	0.0274 (.0044)	0.030 (.001)	0.024	0.030 (.006)	0.0267 (.029)	0.030 (.003)	7
110	0,0200 (.0064)	0.0195	0.01/*	0.022 (.004)*	0.0230 (.005)	0.020*	
111	0.0182 (.0014)	0.01/0 (.0003)	0.014	0.018 (.003)	0.0161 (.0019)	0.017(.001)	
112	0.0128 (.0020)	0.0085 (.0001)	0.010	0.014 (.003)*	0.0120 (.0010)	0.012 (.001)	6,9
113	0.0129 (.0020)	0.0000	0.009		0.0122 ( 020)	0.011#	9
114 175	10.0129 (.0040)	0.0095 ( 0014)	0.014	0 0104 ( 002)"	0.0122 (.000)	0.0110 ( 0014)	
116	0.0103 (.0008)	0.0097*	0.011	0.0104 (4002)	0.0122 ( 051)	0.0109 (.0010)	å
	V.UIII (.00/5)	0.0027	0.011		A.ATTE (.021)	A * ATT	ļ
Total	99.9999	99.48307	99.98548	99.9982	100.019	100.0493	
EAiYi	9489.581	9438,137	9483.413	9488.697	9489.025	9492.555	
XL	94.896	94.872	94,848	94.889	94.872	94.879	

TABLE A1 EVALUATED CHAIN YIELDS FROM ²³⁵U THERMAL NEUTRON FISSION - LIGHT MASSES

* Interpolated yield

- 1. The ENDF/B value is low because the error assignment favors a recent measurement of the yield of 13-s ''Ga. The earlier measurement using longer-lived isobars are probably as accurate, so the higher value is preferred.
- 2. The Crouch yields for Kr are 2 1/2% lower and not included in the average.
- 3. The Crouch yields are high due to the inclusion of early mass spectrometric data [86]. These have isotopic abundances very different from later results (summarized in [7]) and probably should not be used. At mass 88 the listed input does not give the weighted and simple means.
- 4. There are three mass spectrometric measurements of the abundances of Mo isotopes (masses 97, 98, 100). These agree to better than 1% [7] and the ratios have therefore been retained in the recommended yields.
- 5. The ENDF/B value is chosen since it is the only one based on a complete set of data.
- 6. The recommended value is based on yields listed in the ENDF/B library, but excludes one yield (used in the ENDF/B evaluation) that is 25% lower than the recommended value.
- 7. The ENDF/B evaluation uses an unpublished yield that is assigned a high weight; the Lammer and Eder value is also smaller than most measured yields. Both are omitted in taking the average.
- 8. The Crouch evaluation assigns low errors (and hence greater weight) to a 1957 measurement while the ENDF/B evaluation favors a 1970 measurement. The latter set is preferred.
- 9. The Lammer and Eder and ENDF/B evaluations use a preliminary set of mass spectrometric yields that have not been corrected for capture in ¹¹³Cd. It is recommended that a smooth curve be assumed until final values are available.

TABLE A2									
EVALU	JATED	CHAI	N Y	IELD	s	FROM	²³⁵ U		
THERMAL	NEUTI	RON F	ISS	ION	-	HEAVY	MASSES		

Mass	Meek & Rider	Crouch	Lammer	Walker	ENDF/B	Recommended	Note
			£				{
			Eder				<u> </u>
117	.0161 (.0025)	.0100 (.0002)	.011	.0105 (.0020)*	.0103 (.0008)	.011*	1
118	.0147 (.0047)	.0100 (.0002)	.011	.0105 (.0020)*	.0116 (.0027)	.011*	ī
119	.0126 (.0040)	.0110 (.0002)	.012	.0105 (.0020)*	.0113 (.0025)	.011*	1
120	.0131 (.0042)	.0110 (.0002)	.013	.011 (.002)*	.0118 (.0025)	.012*	, 1
121	.0178 (.0014)	.0111 (.0004)	.014	.0130 (.0017)	.0144 (.0008)	.014 (.001)	11
122	.0151 (.0048)	.0130 (.0003)	.015	.013 (.003)*	.0143 (.0044)	.014*	1
123	.0239 (.0019)	.0140 (.0003)	.0164	.016 (.001)	.0158 (.0009)	.016 (.001)	1
124	01/8 (.0114)	0206 ( 0027)	.024	.020 (.004)*		030 / 002)	1 <b>4</b> 5 <b>1</b>
126	0523 (0334)	A 100 ( 002)	.063	053 (010)*	0563 (.031)	.055*	<u>1</u>
127	139 (.005)	0.250 (.005)	0.11	0.124 (.010)	125 (.008)	.125 (.008)	ĩ
128	.402 (.064)	0.500 (.010)	0.36	0.34 (.03)	.348 (.014)	.35 (.02)	1
129	.853 (.130)	1.00 (.02)	0.64	0.88 (.30)	.653 (.051)	.65 (.15)	1,2
130	2.003 (.64)	2.00 (.04)	2.00	1.7 (0.5)	1.442 (.157)	1.7 (.3)	1,3
131	2.771 (.028)	2.85 (.03)	2.82	2.80 (.07)	2.832 (.056)	2.82 (.07)	
132	4.124 (.041)	4.26 (.04)	4.20	4.17 (.09)	4.231 (.060)	4.20 (.09)	i i
133	6.760 (.068)	6.72 (.03)	6.73	6.79 (.16)	6.771 (.068)	6.75 (.16)	1
134	7.187 (.072)	7.76 (.08)	7.67	7.61 (.17)	7.683 (.107)	( 1.05 ( .1/)	
	6.720 (.268)	6.45 (.13)	6.33	6.00 (.10)	6 292 ( 125)	6 18 ( 14)	4
130	6 224 ( 062)	6 27 ( 06)	6.26	6.24 (.16)	6.277 (.031)	6.26 (.16)	
138	6.741 (.135)	6.80 (.17)	6.82	6.76 (.18)	6.827 (.095)	6.80 (.17)	ł
139	6.583 (.263)	6.44 (.13)	6.55	6.53 (.12)	6.477 (.091)	6.50 (.12)	
140	6.316 (.063)	6.32 (.03)	6.37	6.36 (.14)	6.320 (.063)	6.36 (.06)	5
141	5.862 (.234)	5.70 (.17)	5.85	5.87 (.12)	5.575 (.118)	5.82 (.06)	5
142	5.952 (.060)	5.86 (.06)	5.91	5.96 (.13)	5.935 (.084)	5.87 (.06)	5
143	5.987 (.060)	5.89 (.12)	5.92	5.95 (.08)	5.9/8 (.042)	5.95 (.08)	<u>د</u>
144	13.444 (.054)	5.42 (.11)	5.44	3.43 (.10)	3.458 (.055)	3.39 (.00)	3
145	2 996 ( 030)	2 95 ( 06)	2 96	2.98 ( 04)	2 995 ( 021)	2,97 (.04)	
147	2.253 (.022)	2.17 (.08)	2.22	2.26 (.04)	2,274 (.046)	2.25 (.04)	
148	1.689 (.008)	1.69 (.02)	1.67	1.68 (.03)	1.694 (.012)	1.68 (.03)	
149	1.070 (.011)	1.01 (.06)	1.05	1.08 (.07)	1.091 (.022)	1.07 (.06)	}
150	0.649 (.006)	0.637 (.006)	0.644	0.652 (.009)	0.648 (.007)	0.648 (.009)	L
151	0.435 (.009)	0.410 (.008)	0.407	0.419 (.027)	0.422 (.006)	0.420 (.027)	6
152	0.265 (.003)	0.234 (.011)	0.262	0.268 (.017)	0.272 (.016)	0.270 (.017)	6
153	0.163 (.006)	0.150 (.005)	0.163	0.167 (.011)	0.163 (.007)	0.164 (.010)	1 -
155	0332 (.0013)	0052 (.005)	.072	0321 ( 0022)	0331 (.0019)	.0325 (.002)	lí
156	.0133 (.0011)	.0156 (.0003)	.014	.0131 (.0007)	.0136 (.0011)	.0133 (.0011)	1
157	.00642 (.00050)	.00677 (.00041)	.0062	.0061 (.0004)	.0065 (.00072)	.0064 (.00072)	1
158	.00428 (.00035)	.0020 (.0003)	.0031	.0031 (.0006)	.0032 (.00086)	.0031 (.0006)	1
159	.00109 (.00009)	.00101 (.00003)	.00105	.0010 (.0001)	.00102 (.00008)	.00102 (.00008)	11
160	.00033 (64%)		.00035*		.00035 (45%)	.00035*	1
161	.000084 (8%)	.000080 (7%)	.00009	.0007	.000088 (8%)	.000088 (8%)	1
>161	.000040 (32%)	~.00004	~.00004	4	.000040 (328)	.000040"	L
ΣΥ,	99.9996	100.4981	100.0022	99.9969	99.9975	99.705	
Σ <b>Α.</b> Υ.	13867.711	13924.239	13866.384	13868.302	13871.689	13828.315	1
<u> </u>	138 678	138.552	138,661	138.687	138.720	138.693	1
н	120.010		130.001	230.007	1001/20		
$\vec{v}$	2.426	2.576	2.491	2.424	2.408	2.428	8

- 1. The Crouch evaluation assigns low errors to estimated yields of Farrar and Tomlinson [87].
- 2. The Walker value is not based on all the measurements available.
- 3. The ENDF/B evaluation uses only a 1973 measurement, while the others are based only on earlier measurements. The recommended yield uses all data, as shown in Table A9.
- 4. The recommended value of Crouch is greater than any listed yield; the ENDF/B value is also higher than listed measured yields, but lower than 3 estimated values. The recommended value retains the mass spectrometric yield ratio to other Xe isotopes (131, 132, 134).
- 5. The evaluated yields from Table 2 (section 3) are used.
- 6. The ratio of ¹⁵¹Sm to ¹⁵²Sm yields is affected by capture in ¹⁵¹Sm in the results of Lisman et al. [15] and after correction still differs from other measurements (summarized in [7]). The Walker and ENDF/B evaluations take this into account and are used to obtain the recommended yield.
- 7. The Walker and ENDF/B yields are preferred because the very low  154 Sm value of [15] is omitted in the evaluation.
- 8. The value of  $\tilde{v}$  recommended by Hanna et al [44] is 2.4229 ± 0.0066. Only the evaluated yields of Maek and Rider and Wakker, and the recommended yields give  $\tilde{v}$  values that agree within the error limits. The variation in  $\tilde{v}$  is mainly due to the very different yield values used for masses 129 to 130.

				2.0			
Мавв	Meek & Rider	Crouch	Lammer & Eder	Walker	ENDF/B	Recommended	Note
<73	0,000043		~0.00015*		0.800107	0.00013*	
73	0,00019 (32%)		0.00033*	I I	0.00027 (32%)	0.0003*	
74	0.00064 (32%)		0.0009*		0.00053 (32%)	0.0009*	
75	0.0023 (32%)		0.0023*	0.010	0.00085 (45%)	0,0023*	
76	0.0068 (64%)		0.0068*		0,0062 (32%)	0.0065*	
77	0.0177 (.0028)	0.020 (.003)	0.02	0.020 (.004)	0.0178 (.0075)	0.019 (.005)	•
78	0,098 (.031)	0.04*	0.04*	0.060 (.012)*	0,056(.017)	0.06	1
/9	0.186 (.059)	0.05*	0.09*	0.10(.05)	0.132(.007)	0.10"	1
	0.356 ( 057)	0 33 ( 04)	0.33	0.34 (.04)	0.316 (.049)	0.33 (.04)	· · · · · · · · · · · · · · · · · · ·
82	0.702 (.225)	0.61*	0.60*	0.60 (.12)*	0.562 (.171)	0.60*	
83	1.008 (.020)	1.09 (.04)	1.023	1.00 (.02)	1.017 (.010)	1.013 (.020)	
84	1.689 (.034)	1.81 (.07)	1.69	1.66 (.04)	1.702 (.034)	1.68 (.04)	
85	2.213 (.044)	2.32 (.16)	2.19	2.18 (.05)	2.197 (.016)	2.19 (.05)	
86	2.890 (.058)	3.06 (.12)	2.86	2,80 (.06)	2.870 (.081)	2.84 (.06)	
87	3,990 (.080)	4.18 (.29)	4.01	3.98 (.09)	4,004 (.113)	4.00 (.09)	
88	5.561 (.112)	5.47 (.11)	5,54	5.53 (.09)	5.498 (.154)	5.52 (.09)	
89	7.186 (.29)	6.12 (.30)	6.41	6.33 (10)	6.267 (.174)	6.33 (.10)	
90	6.481 (.129)	6.33 (.25)	6.88	6.81 (.10)	6.804 (.195)	6.83 (.10)	2
91	6.421 (.064)	6.56 (.13)	6.52	6.49 (.08)	6,521 (,131)	6.51 (.08)	
92	6.423 (.064)	6.66 (.20)	6.65	6.67 (.09)	6.61/ (.131)	6.64 (.09)	
93	6.838 (.068)	7.06 (.21)	7.04	7.05 (.09)	(7.006 (.139)	7.04 (.09)	
94	6.541 (.065)	6.80 (.21)	6.81	6.75 (.09)	6.810 (.133)	(5.79(.09))	
95	6.19/ (.124)	6.27 (.31)	0.41	5 56 / DQ1	5 696 7 1121	5 71 / 091	· · · · · · · · · · · · · · · · · · ·
90	5.520 (.055)	5./8 (.1/)	5.75	5 36 ( 09)	5 450 ( 077)	5.40(.09)	3
97	5 134 / 102)	5 24 ( 37)	5.14	5 10 (.08)	5.159 (.145)	5.13 (.08)	
90	5 023 ( 102)	5 08 (.15)	4.89	5.01 (.10)	4.969 (.283)	4.99 (.10)	
100	4 369 (.087)	4 50 (.72)	4.38	4.36 (.08)	4,412 (.124)	4.38 (.08)	
	3,189 ( 063)	3.10 7.095		3.21 (.08)	3.224 (.090)	3.21 (.08)	
102	2.434 (.048)	2.31 (.07)	2.42	2.44 (.08)	2.453 (.069)	2.44 (.08)	
103	1.796 (0.72)	1.61 (.23)	1.60	1.8 (.3)	1.702 (.068)	1.70 (.10)	
104	1.023 (.020)	1.00 (.03)	1.02	1.030 (.033)	1.043 (.029)	1.03 (.03)	
105	0.414 (.132)	0.52*	0.54*	0.53 (.10)	0.506 (.111)	0.53*	
106	0.256 (.005)	0.262 (.008)	0.255	0.253 (.006)	0.257 (.010)	0.255 (.010)	
107	0.118 (.038)	0.105*	0.12*	0.130 (.026)*	0.122 (.037)	0.12*	
108	0.069 (.022)	0.087*	0.065*	0.070 (.014)*	0.066 (.020)	0.0/0*	
109	0.039 (.006)	0.052 (.008)	0,04	0.047 (.005)	0.045 (.004)	0.045 (.005)	
110	0.029 (.019)	0.032	0.03*	0.029 (.006)*	0.02/ (.008)	0.030-	
111		0.020 (.0014	0.021			0.021 (.002)	
175	0.015 (.002)	10.014#	0.015#	0.015 (.001)	0.014 (.004)	0 015*	
113	0.020 (.000)	0.014	0.020*	0.014 ( 003)*	0.013 (.004)	0.015*	
114	0.0215(.008)	0.019 (.002)	0.021	0.017 (.003)	0.020 (.003)	0.019 (.003)	
٤y _i	100.00161	100.319	100.0055	100.003	100,1318	100.2781	
EAiyi	9332,143	9366.964	9338,181	9339.700	9351.917	9359.004	
Ā,	93.320	93.372	93.377	93.394	93.396	93.360	

TABLE A3 EVALUATED CHAIN YIELDS FROM ²³³U THERMAL NEUTRON FISSION - LIGHT MASSES

1. The recommended yields follow the curve through the complementary heavy masses as indicated in Fig. 1 of [7].

2. The Crouch value is very low because of the large weights assigned to mass spectrometric measurements that give ⁸ ⁶Sr/⁹ ⁵Sr yield ratios that are very different from other measurements (as summarized in [7]). The recommended yield is the average of the latter.

3. In the Crouch evaluation the listed yields and weights do not give the weighted mean. The most heavily weighted value may be a misprint.

TABLE A4								
EVAL	UATED	CHAIN	YIELDS	S FROM	²³³ U			
THERMAL	NEUTH	RON FI	SSION -	- HEAVY	MASSES			

Mass	Meek & Rider	Crouch	Lammer	Walker	ENDF/B	Recommended	Note
i			Eder		, 		L
116 117 118 119 120 121 122 122 123 124	.010 (.006) .028 (.004) .030 (.005) .030 (.005) .033 (.005) .045 (.029) .045 (.029) .040 (.013) .065 (.010)	.0154* .015 (.001) .015 (.001) .015 (.001) .017 (.001) .020 (.003) .019 (.001) .024* .031 (.002)	.021* .022 .022 .023 .025 .027* .030 .038* .050	.014 (.003)* .014 (.003) .0145 (.003) .015 (.003) .016 (.003) .018 (.004) .025 (.005)* .037 (.007)* .060 (.012)*	.013 (.004) .013 (.002) .013 (.002) .014 (.002) .015 (.002) .017 (.010) .024 (.006) .036 (.008) .057 (.018)	.015* .015* .015* .015* .017* .018 (.003) .024* .036*	1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1
125 126 127 128 129	0.114 (.009) 0.215 (.034) 0.59 (.10) 1.046 (.335) 1.694 (.542) 2.326 (.744)	.116 (.013) .262* .59 (.09) 1.04* 1.61* 1.61*	0.110 0.18# 0.50* 1.00* 1.56* 2.40*	0.116 (.013) 0.26 (.05)* 0.62 (.12) 1.00 (.20)* 1.70 (.34)* 2.50 (.50)*	0.112 (.018) 0.247 (.079) 0.678 (.108) 0.947 (.303) 1.612 (.366) 2.373 (.759)	0.114 (.015) 0.26* 0.65 (.10) 1.00* 1.60* 2.40*	
131 132 133 134 135	3.505 (.070) 4.835 (.096) 5.958 (.060) 6.154 (.123) 6.098 (.244)	3.51 (.07) 4.81 (.29) 5.88 (.12) 6.14 (.18) 5.81 (.18)	3.54 4.84 6.03 6.15 6.27	3.53 (.08) 4.82 (.11) 5.99 (.21) 6.14 (.14) 6.21 (.15)	3.502 (.049) 4.803 (.096) 6.040 (.085) 6.105 (.086) 6.259 (.357)	3.52 (.08) 4.82 (.11) 6.02 (.21) 6.13 (.14) 6.24 (.15)	
136 137 138 139 140	7.386 (.295) 6.560 (.066) 6.372 (.127) 6.092 (.244) 6.417 (.128)	6.89 (.34) 6.12 (.18) 5.96 (.18) 6.20 (.18) 6.32 (.19)	6.82 6.85 6.00 6.34 6.45	6.88 (.16) 6.76 (.16) 5.84 (.14) 6.41 (.14) 6.39 (.12)	6.900 (.552) 6.798 (.095) 5.890 (.165) 6.441 (.258) 6.452 (.367)	6.87 (.16) 6.80 (.16) 5.92 (.16) 6.40 (.20) 6.43 (.12)	2 3
141 142 143 144 145	5.918 (.119) 6.579 (.132) 5.953 (.060) 4.685 (.047) 3.412 (.034)	6.16 (.37) 6.61 (.26) 5.83 (.18) 4.52 (.18) 3.39 (.10)	6.56 6.61 5.88 4.64 3.39	6.62 (.50) 6.60 (.12) 5.85 (.10) 4.62 (.09) 3.38 (.06)	6.603 (.264) 6.643 (.171) 5.885 (.082) 4.664 (.065) 3.375 (.047)	6.60 (.50) 6.61 (.12) 5.86 (.10) 4.61 (.09) 3.38 (.06)	3 3
146 147 148 149 150	2.547 (.025) 1.869 (.374) 1.285 (.013) 0.766 (.023) (0.532 (.021)	2.46 (.07) 1.82 (.11) 1.24 (.05) 0.773 (.02) 0.503 (.010)	2.53 1.80 1.30 0.76 0.501	2.55 (.04) 1.70 (.05) 1.30 (.022) 0.766 (.021) 0.508 (.020)	2.548 (.036) 1.755 (.070) 1.288 (.018) 0.771 (.044) 0.502 (.010)	2.53 (.04) 1.76 (.07) 1.28 (.02) 0.77 (.02) 0.503 (.020)	
151 152 153 154 155	0.338 (.067) 0.193 (.038) 0.126 (.020) .047 (.002) .030 (.019)	0.338 (.010) 0.198 (.008) .099 (.013) .046 (.002) .0231*	0.32 0.22 0.107 .0449 .026	0.314 (.008) 0.213 (.006) 0.105 (.005) .0456 (.0012) .023 (.005)*	0.324 (.013) 0.209 (.012) 0.109 (.009) .046 (.003) .022 (.007)	0.314 (.020) 0.214 (.010) 0.107 (.009) .046 (.002) .023*	4
156 157 158 159 160 161 >161	.012 (.002) .0077 (.0012) .0014 (.0009) .012 (.008) .0029 (32%) .00013 (16%) .000029 (32%)	.0114 (.0011) .0067 (.0007) .00235* .00091 (10%) .00031* .00012 (10%) 	.012 .0072 .0024* .00091 .00035* .00012	.0116 (.0003) .0065 (.0005) .004	.0119 (.0010) .0069 (.0008) .00127 (.00057) .00091 (.0007) .00028 (32%) .00013 (8%) .000026	.0117 (.0010) .0068 (.0007) .0024* .00091 (.0007) .0003* .00012 (10%) ~.00003*	
Σy _i Ea _i y _i	99.9999 13812.626	97.8613 13518.870	100.0099 13817.286	9 <b>9.9</b> 962 13811.350	100.1255 13832.340	100.0163 13815.602	
ж _н	138.126	138.143	138.159	138.119	138.151	138.134	
Ţ,	2.554	2.485	2.464	2.487	2.453	2.505	5

[†]The recommended yields do not take the Crouch values into account. These sum to only 97.9% suggesting the possibility of renormalization. However, differences from the other sets are not consistent, since about half the 1.4% difference occurs for the Cs isotopes, masses 133, 135 and 137.

- The Crouch and Lammer and Eder evaluations use the mass spectrometric measurements of relative Sn yields by de Laeter and Thode [43]. These differ markedly from radiometric yields, and their use is not recommended until they are confirmed by additional measurements.
- 2. The mass spectrometric yield ratio of 137 to 133 is retained in the recommended yields.
- 3. The mass spectrometric Ce isotopic abundances are retained (masses 140, 142, 144).
- 4. The yield ratio of Walker for ¹⁵¹Sm and ¹⁵²Sm is retained in the recommended yields for the reasons given in note 6 for Table A2.
- 5. The value of  $\tilde{\nu}$  recommended by Hanna et al.[44] is 2.4866 i 0.0069. The value given by the recommended set differs by less than 3 standard deviations. This is considered acceptable in view of the large uncertainties in the yields between masses 116 and 130.

	TABLE A5							
EVALU	ATED C	HAIN	YIELDS	F'ROM	²³⁹ Pu			
THERMAL	NEUTRO	N FIS	SION -	LIGHT	MASSES			

Mass	Meek & Rider	Crouch	Lammer & Eder	Walker	ENDF/B	Recommended [†]	Note
<73	0.00017			+	0.00016 (45%)	0.00016*	
73	0.00025 (32%)	0.00026*	0.00025*		0.00024 (16%)	0.00025*	
74	0.00060 (32%)	0.00059*	0.00062*	0.004	0.00059 (32%)	0.0006*	
75	0.0014 (.0004)	0.0013*	0,0016*		0.0014 (.0004)	0.0014*	
76	0.031 (.0020)	0.003*	0.0035*		0.0031 (.0014)	0.0033*	
77	0.0086 (.0007)	0.0071 (.0007)	0.0075	0.0073 (.0015)	0.0071 (.0011)	0.0073 (.001)	
78	0.029 (.002)	0,026 (.003)	0.028	0.025 (.005)	0.025 (.004)	0.026 (.004)	
79	0.025 (.008)	0.036*	0.06*	0.050 (.010)*	0.050 (.002)	0.05*	
80	0.048 (.031)	0.08*	0,11*	0.12 (.02)*	0.121 (.019)	0.11*	
81	0.182 (,029)	0.182 (.018)	0.186	0.18 (.02)	0.184 (.029)	0.183 (.020)	
82	0.167 (.107)	0.24*	0.24*	0.22 (.04)*	0.224 (.036)	0.23*	
83	0.293 (.006)	0.295 (.009)	0,298	0.295 (.007)	0.297 (.006)	0.297 (.007)	
84	0.470 (.009)	0.478 (.019)	0,482	0.477 (.011)	0.481 (.013)	0.482 (.011)	
85	0.601 (.012)	0.559 (.034)	0.566	0.558 (.013)	0.565 (.015)	0.503 (.013)	
30	0.746 (.015)	0.17 (.00)	0.764	0.758 (.017)	0.761 (.021)	0.761 (.020)	1
0/	1 251 ( 027)	1 26 1 001	0,960	1 37 ( 022)	1 272 / 027)	1 375 / 021	-
80	1.551 (.027)	1.50 (.05)	1.305	1.37 (.02)	1 722 ( 025)	1 74 (04)	
90	2 129 ( 042)	2.09 ( 10)	2.13	2 11 / 03	2 124 (042)		
	2 440 ( 049)	2 47 ( 10)	2 51	2 54 ( 05)	2 521 ( 035)	2 54 ( 05)	
92	2 937 ( 059)	3 01 ( 06)	3.05	3 06 ( 04)	3 036 (.085)	3.05 (.04)	
93	3,793 (.076)	3 91 ( 08)	3.92	3 92 (.05)	3.935 (.079)	3.92 (.05)	
94	4.311 (.086)	A A5 ( 09)	4 48	4.45 (.06)	4.466 (.089)	4.47 (.06)	
95	4.912 (.098)	4 90 ( 025)	5.07	4.98 (.08)	4.979 (.139)	5.01 (.08)	
96	4 950 (.099)	5 08 ( 11)	5 12	5,12 (.07)	5,132 (,102)	5.12 (.07)	
97	5.601 (.224)	5 54 (.17)	5.70	5.58 (.10)	5.737 (.229)	5.59 (.10)	2
98	5.725 (.458)	5.59 (.56)	5.93	5.81 (10)	5.914 (.166)	5.88 (.10)	
99	6.456 (.129)	6.20 (.19)	6.33	6.10 (.36)	6.187 (.353)	6.32 (.20)	3
100	6.898 (.552)	6.74 (.67)	7.16	7.00 (.12)	7.154 (.572)	7,10 (.12)	
101	6.061 (.242)	6.05 (.48)	6.01	6.04 (.19)	5,993 (.168)	6.01 (.19)	4
102	6.108 (.244)	6.00 (.42)	6.09	6.15 (.19)	6.138 (.246)	6.13 (.19)	4
103	6.998 (.280)	5,51 (.39)	5.86	5.94 (.29)	5.818 (.116)	6.95 (.29)	5
104	6.056 (.242)	5.99 (.54)	6.03	6.10 (.19)	6.079 (.243)	6.07 (.19)	4
105	5.409 (.222)	5.47 (.55)	5,47	5.47 (.16)	5.291 (.423)	5.47 (.16)	
106	4.271 (.170)	4.34 (.17)	4.64	4.45 (.22)	4,344 (,174)	4.48 (.22)	4
107	3.06 (.98)	2.70*	3.3*	3.5 (.7)*	3.374 (.540)	3.5*	
108	2.53 (.81)	1.70*	2.0*	2.3 (.5)*	2.307 (.738)	2.3	0
109	1.385 (.111)	1.08 (.05)	1.13	1.3 (.2)	1.424 (.082)	1.3 (.2)	
110	0.74 (.40)	0.057 ( 0.17)		10.03 (.13)*	0.034 (.209)	0.02-	<u> </u>
113		10.407 (.013)	0.11	0.20 (.01)	0.207 (.024)		{
113		0.094 (.004)	0.65	0.11 (.02)			
114	0.065 ( 035)		0.03		0.079(.031)	0.072 (.010)	
115		0.032	0.036	0 038 ( 002)	0 038 (.003)	0.037 (.003)	
116	0.037 (.024)	0.038*	0.035*	10.036 (.007)*	0.036 (.012)	10.036*	†
117	0.036 (.011)	0.039*	0.035	0.035 (.007)*	0.035 (.006)	0.035*	1
118	0.0350 (.0112)	0.039*	0.035	0.035 (.007)*	0.035 (.011)	0.035*	
					4	+	ļ
	100 0000	06 6675	00 0005	100 0022	100 0356	1 101 4030	ł
² Yi	100.0007	20.00/2	23,3390	100.0033	1.00.0000		4
ΣAjY	9896,230	9543.167	9881.418	9890.598	9891.228	10031.897	ł
Ă.	99.003	98,732	98.815	98.902	98.877	98.931	1

The recommended yields do not take the Crouch values into account because of the large renormalization required. The difference appears, in part, to be due to the use of yields attributed to Farrar and Tomlinson [91] that are about 4% less than the values in their paper.

- 1. The mass spectrometric yield ratio of ⁸⁵Rb to ⁸⁷Rb [7] normalized to the mass 85 yield is used to obtain the recommended yield.
- The ENDF/B value is not consistent with the listed yields of Meek and Rider [5] [9]. The recommended yield is the average of the Meek and Rider and Walker values.
- 3. The recommended yield is taken from section 3.5.4.
- 4. The uncertainties in the mass spectrometric Ru yields, particularly for ¹⁰¹Ru, are discussed in section 3.5.4. The evidence favoring a revision of the evaluated yields is not considered conclusive.
- The γ-spectrometric yields are assumed correct (section 3.5.4). The recommended yield is 1.17 times the Walker value.
- 6. The estimated yields of Walker are used since these give better agreement between the calculated and recommended  $\vec{\nu}$  values.

TABLE AG								
EVALU	JATED	CHAI	N YIEI	DS	FROM	²³⁹ Pu		
THERMAL	NEUTH	RON F	ISSION	i	HEAVY	MASSES		

Mass	Meek & Rider	Crouch	Lammer & Eder	Walker	ENDF/B	Recommended [†]	Note
119	.036 (.012)	.040*	.036	.035 (.007)*	.035 (.006)	.035*	
120	.037 (.012)	.042*	.038	.035 (.007)*	.035 (.006)	.036*	
121	.053 (.034)	.042±.006	.041*	.038 (.008)	.038 (.024)	.039 (.008)	
122	.047 (.030)	.049*	.045	.038 (.008)*	.038 (.012)	.040*	
123	.056 (.018)	.058*	.055*	.044 (.009)*	.044 (.007)	.05*	
124	0.158 (.025)	.078*	.075	.055 (.011)*	.055 (.018)	.06*	
125	0.109 (.009)	0.116 (.014)	0.110	0.100 (.015)	0.100 (.016)	.106 (.015)	
126	0.218 (.070)	0.24*	0.23*	0.20 (.04)*	0.200 (.064)	.22*	
127	0.526 (.042)	0.513 (.102)	0.55	0.45 (.09)	0.494 (.079)	.50 (.09)	
128	0.844 (.270)	0.83*	1.0*	0.85 (.17)*	0.849 (.272)	.85*	
129	1.689 (.541)	1.38*	1.65*	1.50 (.30)*	1.501 (.240)	1.5*	
130	2.684 (.859)	2.30*	2.6*	2.50 (.50)*	2.503 (.800)	2.5*	
131 132 133 134 135	3.890 (.078) 5.164 (.103) 6.839 (.068) 7.226 (.145) 7.223 (.144)	3.69 (.07) 5.11 (.15) 6.76 (.33) 7.24 (.22) 7.08 (.23)	3.73 5.25 6.94 7.43 7.48	3.73 (.09) 5.21 (.12) 6.92 (.19) 7.41 (.17) 7.69 (.26)	3.759 (.075) 5.281 (.148) 6.974 (.140) 7.417 (.148) 7.326 (.205)	3.74 (.09) 5.23 (.12) 6.92 (.19) 7.42 (.17) 7.69 (.26)	1 1 1 1 1 1,2
136	6.655 (.532)	6.33 (.63)	6.83	6.47 (.15)	6.723 (.188)	6.47 (.15)	1,2
137	6.535 (.065)	6.48 (.39)	6.62	6.72 (.18)	6.687 (.094)	6.65 (.18)	1
138	5.692 (.114)	5.71 (.28)	5.46	5.74 (.37)	5.718 (.457)	5.73 (.37)	3
139	5.842 (.234)	5.77 (.29)	5.82	5.74 (.22)	5.719 (.458)	5.72 (.30)	4
140	5.504 (.111)	5.57 (.28)	5.53	5.62 (.09)	5.583 (.078)	5.59 (.09)	5
141	5.963 (.238)	5.78 (.23)	5.24	5.27 (.35)	5.340 (.214)	5.34 (.25)	4
142	4.977 (.095)	5.05 (.30)	4.97	5.02 (.08)	5.001 (.070)	5.00 (.08)	5
143	4.460 (.045)	4 42 (.22)	4.48	4.53 (.06)	4.553 (.046)	4.51 (.06)	6
144	3.775 (.075)	3.85 (.23)	3.76	3.81 (.08)	3.833 (.027)	3.80 (.08)	5
145	3.017 (.030)	3.14 (.19)	3.04	3.06 (.03)	3.070 (.031)	3.05 (.03)	6
146	2.481 (.025)	2.52 (.21)	2.49	2.53 (.03)-	2.536 (.025)	2.52 (.03)	7
147	1.947 (.039)	2.07 (.13)	2.09	2.16 (.07)	2.102 (.042)	2.13 (.07)	
148	1.659 (.016)	1.71 (.08)	1.68	1.69 (.03)	1.699 (.017)	1.69 (.08)	
149	1.245 (.025)	1.24 (.08)	1.24	1.30 (.05)	1.282 (.025)	1.29 (.05)	
150	0.998 (.020)	0.97 (.03)	0.97	0.989 (.018)	0.996 (.010)	0.99 (.02)	
151 152 153 154 155	0.765 (.015) 0.576 (.012) 0.384 (.061) 0.275 (.005) 0.207 (.065)	0.791 (.048) 0.575 (.058) 0.385 (.039) 0.26 (.03) 0.216 (.03)	0.76 0.58 0.44 0.273 0.17	0.814 (.031) 0.619 (.023) 0.38 (.01) 0.286 (.011) 0.17 (.02)	0.787 (.011) 0.606 (.017) 0.374 (.030) 0.281 (.006) 0.171 (.027)	0.60 (.03) 0.61 (.02) 0.38 (.01) 0.285 (.010) 0.17 (.02)	7 8 7
156 157 158 159 <u>160</u>	.083 (.007) .076 (.048) .042 (.027) .022 (.007) .012 (.004)	.086 (.011) .075 (.011) .04* .021 (.002) .01*	0.12 .080 .045* .022 .011*	0.120 (.010) .076 (.004) .041 (.008)* .021 (.001)	0.120 (.096) .076 (.006) .041 (.013) .021 (.002) .0096 (.0031)	0.120 (.010) .077 (.006) .04* .021 (.002) .010+	
161 162 163 164 >164	.0045 (32%) .0024 (32%) .00098 (32%) .00039 (64%) .0003	.0047 (12%) .0022* .00106* .00042* ~.0003	.0051 .0025* 	.015	.0050 (.0004) .0024 (.0011) .00096 (32%) .00037 (45%) .00023	.0023* .0010* .0004* .00025*	
ΣΥ _i	100.00020	98.6457	99.9904	99.996	99.9868	99.9380	
ΣΑ _i Υ _i	13812.882	13538.727	13810.271	13820.651	13817.822	13810.519	
Ā _H	138.129	137,246	138.116	138.212	138.196	138.191	
7	2.868	4.022	3.069	2.886	2.927	2.878	9

* The recommended yields do not take the Crouch values into account because of renormalization difficulties.

- The uncertainties in the mass spectrometric Xe yields are discussed in section 3.5.4. The evidence favoring a revision of the evaluated yields is not considered conclusive.
- 2. The sum of the mass 135 and 136 yields of Fickel and Tomlinson [91] agree with that obtained by Lisman et al.[15]. The 135 and 136 yield ratio of the former, as used by Walker, is used to give the recommended yields.
- 3. The Lammer and Eder value is apparently based on the smaller of the discrepant values discussed in section 3.5.4. The recommended value uses the evaluated yields based on the average of the discrepant results.
- 4. Only the ENDF/B evaluation uses all data and is recommended.
- 5. The mass spectrometric yield ratios for Ce are retained. For the recommended yields they are normalized at mass 144.
- 6. As in 5, for Nd.
- 7. The Lammer and Eder evaluation is apparently based on the lower of two isotope dilution measurements of the Sm yield. The recommended value is the average of the Walker and ENDF/B values, both of which use both measurements.
- 8. The high value of Lammer and Eder is not included in the average.
- 9. The value of  $\sqrt[n]{\nu}$  recommended by Hanna et al [44] is 2.8799 ± 0.0090. The good agreement with the recommended yield is largely due to the choice of the high  $\gamma$ -spectrometric yield at mass 103.

Mass	Meek & Rider	Crouch	Walker	ENDF/B	Recommended	Note
<75 75 76 77 78 79 80	0.000026 (32%) 0.000067 (32%) 0.000195 (64%) 0.00052 (.0004) 0.0092 (.008) 0.0116 (.0068) 0.0296 (.0095)	0.00045 (.00007) 0.0086 (.0013) 0.018* 0.034*	0.005 0.0082 (.0005) 0.016 (.003)* 0.033 (.007)*	0.000025 (32%) 0.000066 (32%) 0.00019 (45%) 0.00037 (.00004) 0.0082 (.0007) 0.0164 (.0005) 0.0330 (.0075)	0.00010* 0.00025* 0.0008* 0.0025* 0.0083 (_0007) 0.017* 0.033*	1
81	0.055 (.018)	0.063*	0.065 (.013)*	0.0637 (.0145)	0.064*	
82	0.069 (.044)	0.105*	0.120 (.024)*	0.114 (.026)	0.115*	
83	0.198 (.008)	0.201 (.006)	0.202 (.005)	0.205 (.006)	0.203 (.006)	
84	0.343 (.014)	0.353 (.018)	0.360 (.008)	0.357 (.014)	0.357 (.008)	
85	0.379 (.015)	0.387 (.040)	0.392 (.009)	0.396 (.016)	0.392 (.009)	
86	0.588 (.024)	0.601 (.060)	0.608 (.014)	0.613 (.035)	0.607 (.014)	
87	0.725 (.029)	0.741 (.074)	0.750 (.017)	0,756 (.043)	0.749 (.017)	
88	0.934 (.037)	0.954 (.095)	0.966 (.015)	0.973 (.055)	0.966 (.015)	
89	0.760 (.024)	1.19*	1.20 (.24)*	1.182 (.378)	1.19*	
90	1.498 (.060)	1.53 (.15)	1.55 (.02)	1.555 (.089)	1.55 (.02)	
91 92 93 94 95 96	2.183 (.087) 2.829 (.113) 3.260 (1.30) 3.859 (.154) 4.252 (.170)	$\begin{array}{c} 1.76 (.05) \\ 2.23 (.22) \\ 2.90 (.29) \\ 3.33 (.33) \\ 4.00 (.16) \\ \hline 4.33 (.43) \end{array}$	$\begin{array}{c} 1.64 & (.03) \\ 2.26 & (.03) \\ 2.93 & (.04) \\ 3.37 & (.05) \\ 3.98 & (.09) \\ 4.39 & (.06) \end{array}$	$\begin{array}{c} 1.839 (.074) \\ 2.273 (.128) \\ 2.965 (.169) \\ 3.402 (.194) \\ 3.982 (.227) \\ 4.438 (.252) \end{array}$	1.82 (.03) 2.25 (.03) 2.93 (.04) 3.37 (.05) <u>3.99 (.09)</u> 4 39 (.06)	
97	4.587 (.183)	4.75 (.14)	4.73 (.12)	4.762 (.271)	4.75 (.14)	
98	5.924 (1.896)	5.5*	5.2 (.5)*	5.173 (.828)	5.3*	
99	6.267 (.251)	6.14 (.18)	6.20 (.12)	6.231 (.249)	6.19 (.12)	
100	6.221 (1.991)	6.0*	6.2 (.6)*	6.173 (.988)	6.1*	
101	5.683 (.909)	5.94 (.59)	5.91 (.32)	5.968 (.674)	5.94 (.32)	
102	6.047 (.968)	6.32 (.63)	6.29 (.34)	6.344 (.717)	6.32 (.34)	
103	6.023 (1.927)	6.60*	6.65 (.7)*	6.571 (.742)	6.60*	
104	6.506 (1.041)	6.80 (.68)	6.77 (.37)	6.823 (.771)	6.80 (.37)	
105	5.925 (.948)	6.60*	6.75 (.7)*	6.667 (1.067)	<u>6.67*</u>	
106	5.936 (.475)	6.08 (.61)	6.05 (.33)	6.116 (.489)	6.08 (.40)	
107	4.937 (1.580)	5.15*	5.3 (.8)*	5.245 (1.191)	5.25*	
108	3.950 (1.264)	4.15*	4.0 (.8)*	3.94 (1.26)	4.05*	
109	3.778 (.604)	2.9*	2.5 (.5)*	2.48 (.28)	2.6*	
110	2.172 (1.390)	1.4*	1.2 (.24)*	1.18 (.26)	1.3*	
111	1.017 (.325)	0.49 (.07)	0.55 (.04)	0.51 (.08)	0.51 (.07)	
) 12 113 114 <u>115</u> 116	0.911 (.292) 0.167 (.013) 0.059 (.019) 0.0350 (.0224) 0.0267 (64%)	0.32* 0.147 (.022) 0.065* 0.037* 0.033*	0.28 (.05)* 0.153 (.008) 0.075 (.015)* 0.040 (.010)* 0.030 (.010)*	0.28 (.09) 0.15 (.08) 0.074 (.024) 0.044 (.010) 0.029 (.009)	0.30* 0.15 (.02) 0.075* 0.040*	
117	0.0227 (328)	0.031*	0.026 (.010)*	0.026 (.006)	0.028*	
118	0.0207 (328)	0.030*	0.025 (.010)*	0.025 (.011)	0.027*	
119	0.0197 (328)	0.029*	0.025 (.010)*	0.025 (.006)	0.026*	
£Yi	99,99601	100.2484	99.9992	100.0090	100.1410	+
EA _j y _j	100.651	100-393	100.321	100.298	10048.110	+

TABLE A7 EVALUATED CHAIN YIELDS FROM ²⁴¹Pu THERMAL NEUTRON FISSION - LIGHT MASSES

1. The measured yield at mass 77 is about  $1/20^{th}$  that at mass 78. Since such a large ratio is encountered nowhere else the recommended yields are obtained by extrapolating at rates consistent with other measurements (approximately x 0.3 per mass).

TABLE A8						
EVALU	JATED	CHAIN	YIELDS	FROM	²⁴¹ Pu	
THERMAL	NEUTI	RON FI	SSION -	HEAVY	MASSES	

Mass	Meek & Rider	Crouch	Walker	ENDF/B	Recommended	Note
120 121 122 123 124	.0250 (.0080) .0250 (.0080) .0250 (.0160) .0260 (.0083) .0311 (.0200)	.029* .030* .031* .032* .036*	.025 (.010)* .025 (.010)* .025 (.010)* .027 (.008)* .031 (.006)*	.024 (.008) .024 (.008) .024 (.008) .025 (.008) .025 (.008) .029 (.009)	.026* .026* .027* .028* .032*	
123 126 127 128 129 130	.0930 (.0300) 0.700 (.112) 0.454 (.158) 1.008 (.323) 1.818 (.582) 3.0922 (.062)	0.1* 0.21* 0.41* 0.82* 1.65* 3.14 (.09)	$\begin{array}{c}$	0.164 (.057) 0.164 (.052) 0.354 (.113) 0.773 (.175) 1.637 (.524) 3.131 (.088)	.082*. .085* .18* .38* .80* 1.65* 3.13 (.08)	
132	4.589 (.092)	4.59 (.14)	4.64 (.11)	4.658 (.130)	4.64 (.12)	
133	6.611 (.132)	6.64 (.20)	6.72 (.20)	6.715 (.134)	6.72 (.20)	
134	7.990 (.160)	7.99 (.24)	8.08 (.18)	8.102 (.227)	8.09 (.20)	
135	7.293 (.146)	7.08 (.35)	7.06 (.24)	7.180 (.143)	7.11 (.24)	
136	7.210 (.288)	7.04 (.35)	7.30 (.17)	7.290 (.292)	7.29 (.17)	
137	6.562 (.131)	6.52 (.20)	6.50 (.20)	6.650 (.186)	6.58 (.20)	
138	6.594 (.264)	6.54 (.20)	6.71 (.25)	6.519 (.261)	6.60 (.25)	
139	5.882 (1.882)	6.30*	6.3 (0.6)*	6.195 (.991)	6.3*	
140	5.860 (.117)	5.83 (.17)	5.91 (.11)	5.932 (.166)	5.92 (.14)	
141	4.972 (.199)	4.78 (.14)	4.98 (.08)	4.863 (.136)	4.88 (.10)	
142	4.752 (.190)	4.77 (.14)	4.84 (.07)	4.835 (.135)	4.83 (.10)	
143	4.412 (.088)	4.40 (.13)	4.52 (.06)	4.514 (.090)	4.51 (.08)	
144	4.058 (.081)	4.09 (.08)	4.18 (.06)	4.163 (.083)	4.17 (.08)	
145	3.120 (.062)	3.14 (.06)	3.22 (.04)	3.212 (.064)	3.21 (.06)	
146	2.665 (.053)	2.65 (.05)	2.72 (.04)	2.748 (.055)	2.70 (.05)	
147	2.182 (.087)	2.26 (.07)	2.20 (.06)	2.267 (.045)	2.24 (.06)	
148	1.871 (.037)	1.87 (.04)	1 92 (.03)	1.926 (.053)	1.90 (.04)	
149	1.432 (.006)	1.47 (.04)	1.44 (.04)	1.471 (.029)	1.46 (.04)	
<u>150</u>	1.187 (.024)	1.16 (.05)	1.17 (.04)	1.198 (.034)	1.18 (.04)	
151	0.886 (.071)	0.903 (.054)	0.882 (.024)	0.905 (.025)	0.90 (.03)	
152	0.707 (.028)	0.741 (.030)	0.697 (.019)	0.719 (.020)	0.72 (.02)	
153	0.536 (.172)	0.54 (.02)	0.522 (.022)	0.529 (.015)	0.53 (.02)	
154	0.370 (.014)	0.379 (.019)	0.378 (.010)	0.381 (.030)	0.38 (.02)	
<u>155</u>	0.293 (.094)	0.231 (.020)	0.231 (.022)	0.232 (.008)	0.23 (.01)	
156	0.209 (.067)	0.170 (.009)	0.167 (.005)	0.170 (.027)	0.17 (.01)	
157	0.146 (.093)	0.130 (.007)	0.130 (.006)	0.132 (.008)	0.13 (.01)	
158	0.101 (.064)	.086*	0.090 (.018)*	.087 (.007)	.087*	
159	.068 (.043)	.0462 (.0023)	0.046 (.002)	.047 (.015)	.046 (.002)	
160	.046 (.030)	.024*	0.020 (.004)*	.019 (.002)	.020*	
162 163 164 >164	.0021 (32%) .0021 (32%) .00032 (32%) .0002 (32%)		005	.0038 (.0003) .00095 (.00042) .00030 (.00010) .00019 (.00006)	.0038* .0010* .0003* .0002*	
Σy _i ΣΑ _i y _i Ā _H	100.00012 13864.152 138.641	98.91334 13721.767 138.725	100.0012 13873.600 138.734	99.9755 13872.208 138.756	99.9625 13868.503 138.737	
<del></del>	2.708	2.882	2.945	2.946	2.923	1

1. The value of  $\vec{\nu}$  recommended by Hanna et al. [44] is 2.934 ± 0.012.

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# TABLE A9

235 _U	YIELDS	AT	MASS	130
- U	التواجية ليذابية بتديد	13 J	T. 77. P P S S'1	بي بي بي

	Cumulative	Vields	Chain	
Isobar	Absolute (%)	Fractional	Yields (%)	
stable ¹³⁰ Te	$1.43 \pm 0.18[88]$	∿1.000	1.43 ± 0.20	ł
6.6 min. ¹³⁰ Sb	2.17 ± 0.4[63] 2.2 ± 0.5[89]	0.86 ± 0.04 [*]	$\begin{array}{cccccccccccccccccccccccccccccccccccc$	Weighted mean 1,7 ± 0.3
3.7 min. ¹³⁰ Sn	0.89 ± 0.10[88]	0.50 ± 0.10[90]	$1.8 \pm 0.4$	
* ¹³⁰ Sn does not de ¹³⁰ Sb is 1.00 les less the direct y	ecay to ¹³⁰ Sb so the fr as the direct yield to yield to ¹³⁰ Te [90].	actional cumulative yi 33-min. ¹³⁰ Sb (average	eld of 6.6-min. of [88] [90])	

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### APPENDIX B

### A SURVEY OF FRACTIONAL DIRECT AND CUMULATIVE YIELDS FOR REACTOR APPLICATIONS

As noted in the main text, this survey is limited to masses for which the chain yields are about 1% or greater, and, at each mass, to isobars with direct yields about 5% or greater. The latter restriction ensures that all excluded nuclides have cumulative yields that are negligibly small or essentially unity. In the first case uncertainty in the direct yield will have no significant effect on calculations; in the second, its effect on the cumulative yield will be small compared to that due to the uncertainty in the chain yield.

The main source of data is the yield library of Meek and Rider [9], augmented by unpublished yields listed in other surveys [10] [11] [12] and other recent material. An attempt has been made to report each measurement only once since duplications give the impression of a higher degree of agreement than the data warrant. The commonest ways duplications occur are when preliminary values are listed and then the final values are listed following publication, or when cumulative yields are calculated from measured direct yields and both are listed as though they were independent measurements.

Measurements of delayed neutron emission can be converted to cumulative yields if the fraction of the isobar decaying by neutron emission is known. However, the latter are poorly known and yields based on these measurements do not appear in Table B1.

The data is presented in a format that makes unmeasured and undermeasured yields needed for reactor calculations readily apparent. For each mass, measurements for  235 U,  233 U, and  239 Pu are listed in that order to facilitate the isobaric comparison noted in (4) below.

The validity of listed yields, particularly where there is disagreement, can often be checked using the tests listed below. Here  $c_{ij}$  and  $d_{ij}$  are the cumulative and direct fractional yields, respectively, of the ith isobar of mass j, where i = 1 is the stable isobar at the end of the decay chain.

(1) For each mass  $\sum d_{ij} < 1.00$ 

(2) For any isobar 
$$c_{ij} \approx \sum_{k=i}^{\infty} d_{kj} \approx d_{ij} + c_{(i+1),j}$$

(3) Because the average number of  $\beta$ -decays is smaller for ^{2 3 3}U and ^{2 3 9}Pu than for ^{2 3 5}U,  $c_{ij}({}^{2 3 5}U)$  should be greater than both  $c_{ij}({}^{2 3 3}U)$  and  $c_{ij}({}^{2 3 9}Pu)$  for most isobars.

No evaluation is attempted here. The Meek and Rider evaluation for ENDF/B uses as input both measured yields and calculated yields and by least squares fitting works out a complete set of direct yields. The calculated yields are based on a new set of most probable yields* that take into account the systematic odd-even effect investigated by Amiel and Feldstein [12]. Table Bl will provide a useful check on the ENDF/B values when they become available.

* Prepared by Dr. K. Wolfsberg

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	Stable		Cha'n			Frac	tio	nal Yields	of Radioac	tive	Isobars x 1	00		
Mass	(2)	a	(\$)	b	Cum. Di	rect	b	Cum.	Direct	ь	Cum.	Direct	b	Direct ^C
83	Kr(36)	25	0.533	Br (35)		s (3	e,m 4),g m/g m/g	$\begin{array}{cccc} 64\pm4 & [1]\\ 39 & [2]\\ 42\pm4 & [1]\\ 1.5\pm.1 & [3]\\ >93 & [5]\end{array}$	]64±4 [1 ] ]	) As (33)	80±8 [4] 76±6 [5]		Ge (32)	
		23	1.013				g m/g	39±311 1.6±.1 [3	]	<b>†</b>	40° 442 4 900 AN 440 90 400 400 400 400 400 400	********	 	••••
		49	0.297				g m/g	53±7 [3 .92±.12 [3	1	1				****
84	Kr(36)	25	.986	Br (35)		Ċ	Se 34)	99 [6 97±2 [5	]	As (33) mtg	17±2d [4] 15±1d [5] 40±6 [5]		Ge (32)	
		23	1.68							1		**********************		
		49	0.482							]				
85	Rb (37)	25	1.32	Br (35)			Se 34)	87±9 [6 82±7 [5	]	As (33)			Ge (32)	
		23	2.19											*******
		49	0.563											
86	Kr(36)	25	1.95	Br			Se			As		- 41 49 49 49 44 44 46 46 46 46	Ge	
		23	2.84	(35)			34)			(33)	~		(32)	
		49	0.761											
87	Rb (37)	25	2.55	Kr (36)	14	±1[15]	Br 35)	106 [6 95±4 [1]	]47±12 [8 ]43±11 [9	) Se (34)	46±6       [10]         26±5       [12]         41±7       [13]         25±5       [14]		As (33)	4±2 ^e [13] 1.8±.9 ^e [5]
		23	4.00			]		56±4 [10	]75±20 [8		19±4 [10]			
		49	0.98					83±7 [10	]		33±6 [10]			

#### TABLE B1 FRACTIONAL DIRECT AND CUMULATIVE YIELDS FROM THERMAL NEUTRON FISSION

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<sup>a Fissile isotope; 25 = ²³⁵U, 23 = ²³³U, 49 = ²³⁹Pu
b Radioactive isobar and atomic number, Z, in brackets
c For these isobars the cumulative yield is approximately equal to the direct yield
d 5.3s ³³As. It may be either the ground state or metastable.
e. The yield given in the original paper [13] is an absolute value equal to .04 of the recommended chain yield (2.55%). The same authors later quote their result as .018[5]. Smaller changes also occur for ³⁷Se and ⁸⁶Se; the values listed in [5] are, respectively, 7% greater and less than those in [13].</sup> 

	Stable		Chain					Fra	actic	onal Y	ields	of Rad	lioact	ive	Isobar	3 X 1	.00				
Mass	(Z)	a	(8)	b	с	um.	Dir	ect	d	С	um,	Dir	ect	ь	Cur	n .	Dire	ect	ь	Dir	ect ^c
88	sr(38)	25	3.62	Rb (37)					Kr (36)	104±6	[16]	37±3	[15]	Br (35)			56±23	[9]	Se (34)	24±6e 11±1 13±2	[13] [12] [-14]
		23	5.52		<b>-</b> -							·			47±6	[10]				****	
		49	1.375				[								61±3	[10]	[				
89	Y (39)	25	4.80	Rb (37)			4.7±1. 4.0±.8	6[22] [18]	Kr (36)			69±4	[15]	Br (35)			44±10 48±7	[8] [9]	Se (34)	2.3±.8	[12]
		23	6.33							86±1	[19]				19±9	[10]					
		49	1.74	1						86±1	[19]				63±25	[10]					
90	Sr(38)	25	5.89	Rb			13±1 13±1	[18]	Kr	86±2	[20]	63±8	[15]	Br					Se		
		23	6.83	(37)				-1221	(36)	67±1	[19]			(35)	10±3	[10]			(34)		
		49	2.12		- <b>-</b>				ĺ	64±1	[19]				10±4	[10]					
91	Zr(40)	25	5.93	Sr (38)			3±3	[17]	Rb (37)	93±5	[23]	40±2 39±3	[22] [18]	Kr (36)	59±1	[20]	54±2	[15]	Br (35)	7.5±7	[9]
		23	6.51		98±4	(Av.)		<b></b>	1						33±1 ^d	[19]					
		49	2.54		95±2	(Av.)			1					1	31±1 ^d	[19]					
92	Zr(40)		5.97	Sr (38)		h			Rb (37)			62±16 40±3 55±2	[21] [18] [22]	Kr (36)	31±1	[20]	25±1	[15]	Br (35)	< 3	[9]
		23	6.64						1					1	13±1	[19]					*
		49	3.05	1				•	1	/		+		1	12±1	[19]					
93	Zr(40)	25	6.40	Sr (38)					Rb (37)		<del>.</del>	48±3 49±3	[22] [18]	Kr (36)	7.8±.8	[23]	8.3±.5	[15]	Br (35)		
		23	7.04						1					1	2.3±.1	[19]			i i	' 	
		49	3.92						1			<b></b> -		1	2.1±.1	[19]					
94	Zr(40)	25	6.44	Y (39)	6±2	[68]			Sr (38)					Rb (37)			23±1 27±2	[22] [18]	Kr (36)	1.5±.6 4.0±.3	[23] [15]
		23	6.79					~	1					1							
		49	4.47	1					1		<b></b>			1							

### TABLE B1, continued (All references are for the Appendix B reference list)

a Fissile isotope;  $25 = {}^{235}$ U,  $23 = {}^{233}$ U,  $49 = {}^{239}$ Pu b Radioactive isobar and atomic number, Z, in brackets c For these isobars the cumulative yield is approximately equal to the direct yield d Multiplied by fractional cumulative yield of  91 Sr e See footnote e on preceding page

	Stable		Chain			Fra	ictio	nal Yields	of Radioact	ive	Isobars x 1	.00			
Mass	(Z)	a	(8)	b	Cum.	Direct	b	Cum,	Direct	Ь	Cum.	Direct	b	Direct	c
95	Mo (42)	25	6.50	Zr	99±1 (Av)	.4±.2 [24]	Y		13±6 [25]	Sr			Rþ	10±.5 [2	2]
							Ŷ								.01
		23	6.22	(40)	100±3 (Av)	3.4±.6 [24]	(39)	L		(38)			(37)		
		49	5.01		99±2 (Av)										
96	Zr(40)	25	6.28	Rb		2.0±.1 [22]	Kr			Br			Se		
				(37)			(30)	)  ****		(35)			(24)		
		23	5.71				Į								·
L		49	5.12							Į					
97	Mo (42)	25	6.03	Zr			Y (39)			Sr			Rb (37)	$0.5\pm.1$ [2	2]
							(3)								
			5.40				4								-
		49	5.59				<u> </u>	ļ							
98	Mo(42)	25	5.79	Zr			Y			Sr			Rb		_
		23	5.13	(40)			(39)			(38)			(37)		
		49	5.88												
99	Tc(43)	25	6.13	Mo		6±1 [26]	Nb,m	33±3 [26]		Zr	1.9±1 [26]		Y		
				(42)			(41) q	31±2 [27] 61±11 [26]	30 [28]	(40)			(39)		
		23	4.99												
			6 22			~~~~~	m		29±1[27]		 				
		49	0.32				<u> </u>								
100	MO (42)	25	6.30	Zr			Ŷ			Sr	ane ant ann dar 144 448 479 425 466 478 4		Rb		
		23	4.38	(40)			(39)			(38)			(37)		_
		49	7.10												
101	Ru (44)	25	5.05	Мо			Nb			Zr			Y		
1		23	3.21	(42)			(41)			(40)			(39)		-1
		49	6.01					<b>}</b>					1		-1
102	Ru (44)	25	4.19	Mo	<u> </u>	<b></b> ,	Nb			Zr			Y		┥
		23	2.44	42)			(41)			(40)			(39)		-
}				· /			1			,,			,		-
1	1	49	D.13		}		1								

TABLE B1, continued (All references are for the Appendix B reference list)

a Fissile isotope;  $25 = {}^{235}U$ ,  $23 = {}^{233}U$ ,  $49 = {}^{239}Pu$ 

b Radioactive isobar and atomic number, Z, in brackets c For these isobars the cumulative yield is approximately equal to the direcy yield

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	Stable		Chain			··· ··	Fra	actic	nal Yi	elds	of Rad	lioact	ive	Isobars x 1	00		
Mass	(Z)	a	(8)	b	Cum.	Dir	ect	b	Cu	n.	Dii	rect	b	Cum.	Direct	b	Direct ^C
103	Rh (45)	25	3.12	Tc				Мо			}		Nb			Zr	
		23	1.70	(43)				(42)	<b>_</b>				(41)	**********		(40)	********
		49	6.95					]									
104	Ru (44)	25	1.83	Мо				Nb					Zr			Y	
}		23	1.03	(42)				(41)	[		[		(40)			(39)	
		49	6.07						[								
105	Pd(46)	25	0.927	Ru				TC	76±3	[29]			Мо	98±2 [30]		NĐ	
		23	0.53	(44)				(43)					(42)			(41)	
		41	5.47						72±8	[29]							
106	Pd (46)	25	0.390	Ru				Tc	L	*			Мо	107±9 [30]		Nb	
		23	0.255	(44)		L		(43)					(42)			(41)	
		49	4.48						80±10	[29]							
107	Pd (46)	25	0.17	Ru				TC	33±5	[29]			Мо			Nb	
1		23	0.12	(44)				(43)	L				(42)			(41)	
		49	3.5						41±5	[29]							
129	I(53)	25	0 65	Sb (51)		11±5	[32]	Sn,π 50)g	53±3 36±3 31±1	[32] [32] [34]			In (49)			Cđ (48)	
		23	1.60					]			[		Í				
		49	1.5														
130	Te (52)	25	1.7	Те (52)		3	[28]	Sb,m (51) g			37 20±3 9 14±3	[28] [32] [28] [32]	Sn (50)	50 [28] 52±6 [32]		In (49)	
		23	2.4							_~~~~							
		49	2.5					l									

TABLE B1, continued (All references are for the Appendix B reference list)

a Fissile isotope;  $25 = {}^{235}$ U,  $23 = {}^{233}$ U, 49 = 239Pu b Radioactive isobar and atomic number, 2, in brackets c For these isobars the cumulative yield is approximately equal to the direct yield

	Stable		Chain	1				Fra	actio	nal Yiel	ds	of the	Radi	oact	ive Isc	bars	x 100				
Mass	(Z)	a	(%) (%)	ь	Cu	m.	Dire	ect	b	Cum.		Dire	ect	b	Cur	a .	Dir	ect	b	Dir	ect C
131	Xe (54)	25	2.82	re,m (52) g	16e 15e	[35] [36]	2.5±2.5 11 8±1 51±10 1.5±1.5 4.4±.7 12	5 [ 37] [ 38] [ 39] [ 36] 5 [ 37] [ 39] [ 28]	Sb (51)	64±2 [ 91±9 [	36] 40]	59±14 52	[41] [28]	\$n (50)	28 35 33±6 45±7	[42] [28] [32] [41]			In (49)		
		23	3.52	l m g	98	[43]	23	[38]													
		49	3.74	m			18±5	[38]													
132	Xe (54)	25	4.20	Те (52)	102±5	[Av]	20±2 38±2 2	[41] [44] [45]	Sb (51)	72 [	40]	66±8 49±1 16±2	[41] [44] [45]	Sn (50)	14±4 14±1 20 32±3	[41] [44] [42] [46]	56	[45]	In (49)	25±2	[45]
		23	4.82	Ì			64±4	[44]	į			27±3	[44]		2.5±.5	[44]				******	
		49	5.23	ļ	100±5	[Av]	2	[45]		' 		9±7	[45]				61	[45]		27±5	[45]
133	Cs (55)	25	6.75	I (53)			26 2.5±2.5 2.2±.8 12 25±.8 2±2 2.5±1.4 2	[36] 5[37] [41] [47] [47] [48] [49] 4[50] [45]	Te,m (52)g m+g	71 [	36]	29±4 5±5 28±4 45±7	[41] [37] [41] [45]	<b>Sb</b> (51)	56 42±10	[36] [51]	<b>46</b> 51±5	[41] [45]	Sn (50)	3	[45]
		23	6.02		[		14±1 21±1£	[52] [53]													
		49	6.92				15±3 6	[52] [45]				40±7	[45]				49±5	[45]		5	[45]

TABLE B1, continued (All references are for the Appendix B reference list)

a Fissile isotope, 25 = ²³⁵U, 23 = ²³³U, 49 = ²³⁹Pu b Radioactive isobar and atomic number, Z, in brackets c For these isobars the cumulative yield is approximately equal to the direct yield e About 7% of ¹³¹Sb decays to ¹³¹MTe[39] so that most of the cumulative yield is formed directly

f Revised to 15,5±1 (private communication from HO.Denschlag, 1974)

	Stable		Chain				Fra	ctic	onal Yie	lds	of the	Radi	oact	ive Isc	bars	x 1.00				
Mase	(Z)	a	(\$)	b	Cum.	Dire	ect	b	Cum	ı.	Dire	ect	ь	Cur	a .	Dir	ect	b	Dir	ect
134	Xe (54)	25	7.65	I (53)		13 11±2 12 10±1 12±2 12±1 35±5	[54] [37] [47] [28] [49] [50] [45]	Те (52)	89±1 90	[56] [28]	52±6	[45]	Sb (51)	4.2±.5 3.8	[55] [51]	16	[45]	Sn (50)	1	[45]
		23 49	6.13 7.42			33±2 37±2 34±3	[52] [53] [52]				51+6	 [45]		***						
135 ^d	Cs (55)	25	6.60	Xe	100	3.5	1451	I	96.5		47		Те	50		(47)	1401	Sb	(3)	1451
		23	6.24	(54)	>99	21.0		(53)	79.0		 61		(52)	18		(18)		(51)	<b>{∿0</b> }	
		49	7.69		>99	15.0	***		85.0		61			24		(23)			(~1)	
136	Xe(54)	25	6.18	<b>Xe</b> (54)		21±4 24±18 34±5	[57] [58] [45]	I (53)	65±15	[58]	67±25 21±8 35±5	[57] [48] [45]	Te (52)			10±3 24±4	[57] [45]	Sb (51)	5±3	[45]
		23	6.87				* *******													
		49	6.47			32±5	[45]			4= 10 44	33±3	[45]				25±4	[45]		5±3	[45]
137	Cs (55)	25	6.26	Xe (54)	97.8±.3 [23]	33±5 48±6	(57] [15]	I (53)			53±6 28±6 46±7	[57] [59] [9]	Те (52)			8	[57]	Sb (51)	1	[57]
		23 49	6.80		$90\pm1$ [19] $92\pm1$ [19]				28±3 42±2	[10] [10]				16±4	[10]	******				
138	Ba (56)	25	6.80	Cs (55)		4.7±.2 4.8±.1 14	[23] [60] [45]	Xe (54)			74±8 56±5	(15) [45]	I (53)			17±3 12±3	[45] [59]	Те (52)	4±2	[45]
		23	5.92						83±1	[19]				10±1	[10]	*****				
		49	5.73			22	[45]	]	85±1	[19]	55±7	[45]		14±9	[10]	15±3	[45]		4±2	[45]

TABLE B1, continued (All references are for the Appendix B reference list)

a Fissile isotope;  $25 = {}^{235}$ U,  $23 = {}^{233}$ U,  $49 = {}^{239}$ Pu b Radioactive isobar and atomic number, Z, in brackets c For these isobars the cumulative yield is approximately equal to the direct yield d Recommended fractional yields from Table 4

	Stable		Chain				Fra	actic	onal Yields	of the Rad	loaci	tive Isobars	× 100			
Mass	(Z)	a	(%)	b	Cum.	Dire	ect	b	Cum.	Direct	b	Cum.	Direct	b	Dire	ct
139	La(57)	25	6.50	Ba (56)		1.1±.4 5±5	[23] [69]	Cs (55)		24±2 [22 20±3 [60 18±2 [18	Xe (54)	82±2 [56]	79±7 [15] 13±3 [69]	I (53)	12±5 7±4 2±1	[9] [59] [69]
		23	6.40			8.4±.7	[61]					48±1 [19]			≤11	[10]
		49	5.72									55±1 [19]			5.4±3.4	[10]
140	Ce(58)	25	6.36	Ba (56)		4.6±3.0	[23]	Ся (55)	93±3 [23]	31±2 [22] 33±3 [18]	Xe (54)	60±1 [56]	46±6 [15]	I (53)	3.4±2.1	. [9]
		23	6.43			27±4	[61]					23±1 [19]				
		49	5.59									30±1 [19]				
141	Pr(59)	25	5.82	La (57)		.30±.04 .37±.13	4[62] 3[23]	Ba (56)		<b>26±</b> 5 [23	Cs (55)		55±1 [22] 45±7 [60] 52±4 [18]	Xe (54)	21±2 20±3	[20] [15]
		23	6.60								]				5.1±.3	[19]
		49	5.34					1							7.9±.4	[19]
142	Ce(58)	25	5.87	La (57)		1.7±.4	[23]	Ba (56)			Cs (55)		41±1 [22] 25±7 [60]	Xe (54)	6±5 11±2	[63] [15]
		23	6.61												1.0±.1	[19]
		49	5.00												1.7±.1	[19]
143	Nd(60)	25	5.95	Ce (58)		.53±.03 12±3	8[23] [64]	La (57)	70 [65]		Ba (56)	88±6 [66]		Cs (55)	25±1 17±3 20+3	[22] [60]
		23	5.86					]			1	· · · · · · · · · · · · · · · · · · ·				1101
		49	4.51													
144	Nd (60)	25	5.39	Ce		1.2±1.2	2[67]	La			Ва	78±6 [66]		Cs	5.2 <b>±1.6</b>	[22]
			]	(58)				(57)			(56)			(55)	22±3	[18]
		23	4.61													
		49	3.80													

TABLE B1, continued (All references are for the Appendix B reference list)

a Fissile isotope;  $25 = {}^{235}$ U,  $23 = {}^{233}$ U,  $49 = {}^{239}$ pu b Radioactive isobar and atomic number, Z, in brackets c For these isobars the cumulative yield is approximately equal to the direct yield

	Stable		Chain	Γ		Fra	actic	onal Yields	of Radioact	ive	Isobars x 1	00		
Mass	(Z)	a	(\$) XJETQ	Ъ	Cum.	Direct	b	Cum.	Direct	b	Cum	Direct	ь	Direct ^C
145	Nd (60)	25	3.93	Çe		61 [64]	La			Ва			Ся	7±2 [13]
		23	3.38	(58)			(57)	************		(56)		†	(55)	91206434323
		49	3.05				1		*****					
146	Nd (60)	25	2.97	Pr			Ce			La			Ba	
		23	2.53	(59)			(58)			(57)	[		(56)	
		49	2.52				]							
147	Sm (62)	25	2.25	Pr			Ce	96±5 [17]		La			Ва	
		23	1.76	(59)			(58)			(57)			(56)	
		49	2.13			********								
148	Nd(60)	25	1.68	Pr			Ce			La			Ba	
		23	1.28	(59)			(58)			(57)			(56)	
		49	1.69											
149	Sm (62)	25	1.07	Nđ			Pr			Ce			La	
		23	0.77	(60)			(59)			(58)			(57)	
		49	1.29											

TABLE B1, continued (All references are for the Appendix B reference list)

a Fissile isotope;  $25 \approx 2^{35}$ U,  $23 = 2^{33}$ U,  $49 = 2^{39}$ Pu b Radioactive isobar and atomic number, Z, in brackets c For these isobars the cumulative yield is approximately equal to the direct yield

### APPENDIX C

### MISCELLANEOUS TABLES

Two tables were prepared for the oral presentation which may be of general interest.

Table Cl shows revisions to the ¹⁴⁰Ba yields of Table 2. The investigation was initiated by a letter from M. Lammer noting that the result of Santry and Yaffe [31] required renormalization. The remaining  $\beta$ -counting measurements, with one exception, required corrections for flux depression. The changes increase the weighted mean by 0.8%.

Table C2 is a revised table of the thermal/epithermal counting rates given in Tong et al.[81]. The purpose of the data as originally presented was intended to show that for each irradiation this ratio was constant within experimental error for many radioactive fission products in the peak region.

In Table C2 each measured ratio is divided by the mean ratio for the same irradiation so that the numbers all lie within a few percent of unity and those in the bottom line are, of course, exactly one. These ratios of ratios are then averaged for each nuclide with the average values and their rms deviations listed in the right hand column. Only for mass 140 does the average value deviate from unity significantly. Even here 2 of the 3 measurements differ by appreciably less than the assigned error of 4%.

### TABLE C1

# MASS 140 YIELDS FROM 235U FISSION

	Chain Yiel	đs (%)	
<u>Ref. year</u>	<u>Table 2</u>	<u>Corrected</u>	Comments
27 (1951)	6.17 <u>±</u> .13(.62)	6.51 <u>+</u> .62	Fission chamber; $2\pi\beta$ counting of ¹⁴⁰ Ba + ¹⁴⁰ La. Counting correction by Glen- denin quoted in [30]. Target thickness equivalent to ~ 20 mil foil; no correc- tion for flux lepression, estimated correction (3±0.5)%.
28 (1952)	6.37±.18(.64)	6.55 <u>+</u> .64	Fission chamber; $2\pi\beta$ counting of ¹⁴⁰ Ba + ¹⁴⁰ La. Target 20 mil foil; no correc- tion for flux depression; estimated correction (3±0.5)%.
29 (1953)	6.25±.13(.59)	6.25 <u>+</u> .59	$2\pi\beta$ counting relative to ⁸⁹ Sr absolute yield. Same authors give absolute yields of ⁹⁷ Zn, ⁹⁹ Mo very different from evaluated values so results are questionable.
30 (1954)	6.32 <u>+</u> .24(.58)	6.55 <u>+</u> .58	Flux from m.s. analysis of $B^{10}/B^{11}$ in B monitor; $2\pi\beta$ counting. Correction is for flux depression as calculated by Petruska et al (CJP <u>33</u> ,693).
31 (1960)	6,36 <u>+</u> ,12(.46)	6.68 <u>+</u> ,46	Co monitor; $2\pi\beta$ counting. Renormalized to $\sigma_f(^{235}U)=557b; \sigma_a(^{59}Co)=37.2b$
33 (1968)	6.36±.32(.32)	6.36 <u>+</u> .32	Fission chamber; Ge(Li) $\gamma$ -detector; counted ¹⁴⁰ La $\gamma$ -ray against ¹⁴⁰ Ba ¹⁴⁰ La standard.
34 (1971)	6.29±.14(.25)	6.29 <u>+</u> .25	Fission chamber; Ge(Li) $\gamma$ -detector; counted ¹⁴⁰ La $\gamma$ -ray against irradiated ¹³⁹ La; ¹⁴⁰ La decays of std. determined by $2\pi\beta$ counting.
18 (1971)	6.40 <u>+</u> .11(.26)	6.40 <u>+</u> .26	Fission chamber; Ge(Li) $\gamma$ -detector; counted ¹⁴⁰ La $\gamma$ -rays and obtained absolute disintegration rate using calibrated detector efficiency.
35 (1973)	5.77 <u>±</u> .30	not used	Mica fission track recorder; Ge(Li) $\gamma$ -detector; counted ¹⁴⁰ La standard; weight of fission foil not given; no correction for flux depression.

Weighted means

6.34±0.35(0.05) 6.39±0.37(0.10)

# TABLE C2

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	FOR ²³⁹ Pu FISSION PRODUCTS									
Experiment No.	1	2	3	4	5					
Shield Material	Sm	Cd	Cđ	Sm	Sm	Av				
Kr-85m		0.991				0.991				
Sr-91 Y-91m	1.029 0.994	1.021 1.013	1.01			$1.013 \pm .012$				
sr-92	1.012					1.012				
Zr-97 Nb-97	1.000 1.012	1.013 1.017	0.994 0.994			1.004±.012				
Tc-99m			0.989	1.017		1.003 ±.014				
Ru-103				1.008	0.986	.997 <u>+</u> .011				
Ru-105	1.029	1.019				1.017				
I-131			1.029	1.000	1.021	1.017				
Te-132 I-132	0.987	0.991 1.017	0.989 1.022	1.008 1.000	1.021 1.007	1.004±.015				
I-133	1.000	0,983	1.017	0.992	0.992	0.997±.013				
I-135 Xe-135	1.017 0.982	1.026 0.995	1.006 0.977	0.967	0.986	1.006+.023				
Ba-140 La-140				0.959 0.975	0.979	0.971±.011				
Ce-143	0.997	0,966	1.011	0,983	0.979	0.987±.018				
Nd-147			1.006	0.967	0.986	0.986±.020				
Weighted average	1.000	1.000	1.000	1.000	1.000					
	±.023	±.021	<u>+</u> .029	±.025	<u>+</u> .029					

RELATIVE (THERMAL/SHIELDED) ACTIVITY RATIOS

Review Paper No. 11b

## REVIEW OF FISSION PRODUCT YIELD DATA FOR FAST NEUTRON FISSION

by

J. G. Cuninghame

Chemistry Division, A.E.R.E., Harwell, Didcot, Berks., England.

### ABSTRACT

This paper summarises the current position on fast fission yields, including cumulative and independent yields, the effect of change of neutron energy, 14 MeV yields, and ternary fission. Tables of data from recent yield evaluations are presented and compared, and new data not yet included in evaluations are given. Recommendations are made for measurements needed to fill the many large gaps in the data.

### 1. INTRODUCTION

This paper has been written with the object of presenting a clear picture of the situation today regarding data on fast fission yields of nuclides important to reactor programmes. The method used is to compare all evaluations published since 1968, provided the evaluator has taken as his starting point the original experimental data and not an evaluation of another author, and then add supplementary information obtained by searching the literature from the date of the latest evaluation until the end of June 1973 or taken from contributions made directly to me especially for this meeting.

The following topics are discussed in this paper:-

- 1. Experimental methods used in fast fission yield measurement.
- 2. Evaluation of fast fission yields.
- 3. Chain yields in fast fission.
- 4. Chain yields in 14 MeV fission.
- 5. The effect of neutron energy on fission yields.
- 6. Charge distribution in fast and 14 MeV fission.
- 7. Ternary fission.

The following topics are not included:-

- 1. Evaluation procedures in general.
- 2. Experimental methods in detail.
- 3. Error analysis.

because they are all dealt with in paper 11a. Similarly, the theoretical background has been left to paper 16.

At this point it seems wise to explain how the term 'fast fission" is interpreted in this paper. All fission yields measured in a fast reactor spectrum, and yields of non-thermally fissile nuclides measured in either a thermal or a fast reactor spectrum, are called "fast yields". We should realise that applying this definition could possibly cause errors if there was any substantial energy effect on the yields, since neutron spectra vary from reactor to reactor and in different areas of the same reactor. However, as we shall see, except for yields in the valley and on the wings of the mass yield curve, any such effect has yet to be detected with certainty. In any case, few authors have given details of their energy spectrum except where they are deliberately seeking to examine its effect. Yields measured in mono-energetic neutron fluxes are not included in the data sets, but are discussed separately in the section on energy effect.

The term "14 MeV fission" also requires some explanation. It refers, of course, to fission by neutrons generated by the  3 H(d,n)⁴He reaction at low deuteron energies. The energy of the resulting neutrons depends on the exact conditions of their generation and authors have quoted a range of values around 14.7 MeV for it. All such measurements are usually referred to as 14 MeV fission, as is the case herein.

Table 1 is a list of evaluations, relected in accordance with the conditions stated in the first  $paragra_{ph}$  of this introduction, which I have used in this paper.

# 2. EXPERIMENTAL METHODS USED IN FAST FISSION YIELD MEASUREMENT

### 2.1 General discussion

The main discussion of the different kinds of experimental methods used in the measurement of fission yields is to be found in paper 11a and this section will be confined to a consideration of which of them are most suitable for fast yields.

The chief difference between thermal and fast fission is, of course, that for all practical purposes thermal fission yields have no energy effect because the very high thermal fission cross-section dominates the

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## Table 1

### Evaluations used in this paper

Author	Ref	Date	Nuclides for which sets of recommended values of fission yields are given				Commen ts
			Spontaneous	Thermal	Fast	14 MeV	
von Gunten	59	1969	238 _U 252 _{Cf}		237 _{Np} 241 _{Am}	232 _{Th} 235 _U 238 _U	
Meek & Rider	3	1972		233 _U 235 _U 238 _U 239 _{Pu}	232 _{Th} 235 _U 238 _U 239 _{Pu}	235 _U 238 _Ù	
Sidebotham	7	1972		237 _U 237 _{Np} 240 _{Pu} 242 _{Pu} 241 _{Am}	232 _{Th} 233 _U 234 _U 236 _U 237 _U 237 _{Np} 238 _{Np} 238 _{Pu}		These are NOT evaluated experi- mental yields, but are calculated by methods explained in Section 4 of this paper.
Sidebotham (cont'd)	7	1972			240 _{Pu} 241 _{Pu} 242 _{Pu} 241 _{Am} 243 _{Am} 242 _{Cm}		
Lammer & Eder	2	1973		233 _U 235 _U 239 _{Pu}	²³² Th		
Crouch	1	1973			232 _{Th} 233 _U 235 _U 237 _{NP} 238 _U 239 _{Pu}	231 _{Pa} 232 _{Th} 233 _U 235 _U 237 _{Np} 238 _U 239 _{Pu} 241 _{Pu}	A companion paper, ref. 4, contains Crouch's thermal yield evaluations.
Daróczy, Raics & Nagy	60	1973				238 _U	

situation. Note that this is not quite the same as saying there is no energy effect in thermal reactors, particularly at high temperatures and high burn-up, but such matters are not in the province of this paper.

Apart from the fact that fission in fast reactors occurs over a wide neutron energy range, a further factor influencing experimental methods is that the low fast fission cross-section make very long irradiation periods necessary if we are to achieve a high enough number of fission in a sample so that the yields can be measured mass-spectrometrically. During these long periods conditions within the reactor may change. If we want to study the energy effect with exactitude we must either use mono-energetic neutrons or else neutrons having a very well-defined spectrum which does not change during the irradiation. In practice this usually means either using neutrons from a nuclear reaction (e.g.  ${}^{7}Li(p,n){}^{7}Be$ ) or else from an experimental low power fast reactor. In both cases the flux obtainable is low, thus making mass spectrometric measurements virtually impossible.

## 2.2 Methods commonly used for measuring fast fission yields

### (a) Mass spectrometry

Provided the problems of long irradiation time and possible variation of the neutron spectrum during irradiation can somehow be overcome, mass spectrometry probably remains the most accurate means of measuring fast yields. Unfortunately these provisos are serious and make it difficult to know how to evaluate such yields when carried out in different reactors (see section 3 on evaluation methods). The method has been used for fast yield measurements, however, notably by Davies⁽⁴⁹⁾ in the UK Dounreay Fast Reactor (DFR) and Lisman et al.⁽⁵⁰⁾ in the US reactor EBR I; both of these papers report results on  $235_{\text{U}}$  and  $239_{\text{Pu}}$ . Davies obtained the number of fissions which had occurred in his samples by mass-spectrometric measurements of the consumption of the target material during the irradiation while Lisman et al., who had measured over 80% of the total heavy mass peak, filled in the remaining yields from values in the literature and normalised their values to 100% for the heavy peak. Overall accuracy of fast yields measured in these ways (i.e. including systematic errors) is probably ± 2 - 3%.

### (b) Very precise $\gamma$ -counting

 $\gamma$ -spectrometry by means of a carefully calibrated Ge(Li) detector can be nearly as precise as mass-spectrometry and, provided the number of

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fissions can be determined to a comparably high accuracy, very good absolute fission yields can be obtained. The total number of fissions needed in the sample is much lower than for mass-spectrometry and it is therefore possible to use better defined neutron spectra, such as those from a small critical assembly. A good example of this technique is to be found in the work of Larsen et al.⁽⁵¹⁾ who irradiated their samples in two different assemblies in ZPR-3, each one being a mock-up of a particular loading for EBR II. They determined the number of fissions in their samples by means of mica fission track detectors. The overall accuracy for fast yields measured in this way is probably about  $\pm 3 - 4\%$ .

## (c) $\beta$ -counting techniques used for very low fluxes

It may not be possible to give the sample a sufficiently large total neutron dose even for precise  $\gamma$ -spectrometry, and in such a case the use of carriers followed by  $\beta$ -counting of a thick sample is the only reasonable method of estimating the amount of fission product present. To achieve the best accuracy very stable, low background, 2  $\pi$  proportional counters (background < 1 count/min) must be used and self-absorption curves must be prepared for the nuclides concerned by means of absolutely standardised samples. Provided an accurate method of obtaining the number of fissions in the sample is employed the overall accuracy of the yields can be in the range  $\pm 4 - 8\%$ .

Examples of the use of this procedure are Bowles and Willis' absolute yields for  235 U fissioned in DFR⁽⁵²⁾, Cuninghame, Goodall and Willis' absolute yields for  235 U,  238 U and  239 Pu in DFR⁽⁵³⁾ and the absolute yields for  235 U in mono-energetic neutron fluxes by Cuninghame, Goodall and Willis⁽¹²⁾. Fission chambers were used to measure the number of fissions in the DFR experiments and fission track detectors for the mono-energetic neutron measurements.

Thick source  $\beta$ -counting and  $\gamma$ -counting have also been used by many authors for determining relative fast fission yields by techniques which have been in use for many years.

#### (d) Summary of the methods

Table 2 gives a summary of the methods discussed above for the measurement both of the amount of fission product and the number of fissions, together with my strictly subjective estimates of the overall accuracy (i.e. including all errors) which can reasonably be expected.

# Table 2

# Summary of methods used in fast fission yield measurement

# Part I: Estimation of the amount of fission product present

Name of method	Percentage Accuracy (as a standard deviation)	Comments
Mass spectrometry	2 - 3	Requires large number of fissions in sample : long irradiation in high flux.
Precise Y-counting	3 - 4	Suitable for irradiations in lower flux, e.g. criti- cal assembly. Requires accurate calibration of counter and standards.
Low background thick source β-counting	4 - 8	Source disintegration rate can be low (1 - 2 dis/min) ∴ suitable for very low flux. Requires good self- absorption curves and first rate counter.
Relative yields by $\beta$ or $\gamma$ counting	5 - 20 (depends on accuracy of reference yield)	The classical method. Quick and easy but not accurate.
Part II: Estimation of	total fissions in the	source
Mass-spectrometry of fissile target mater- ial (or of a fission product if the yield is known)	2 - 3	See Part I.
Fission chamber	2 - 4	Needs careful calibration. Can only be used for short (tens of hours) irradia- tions in low fluxes (10 ¹⁰ - 10 ¹¹ n/cm ² /sec) when high accuracy is required.
Fission track detector	Depends on number of tracks counted but not better than 1 - 2	Can be used in quite high fluxes by adjustment of amount of fissile material used. Simple and reliable but tedious to obtain data from them unless automatic counting methods (less accurate than optical counting) is used.

# 2.3 <u>Methods used to examine the effect of neutron energy change in fast</u> <u>fission yields</u>

# (a) Mono-energetic neutrons

It is possible to irradiate samples with different energy bands of neutrons by spinning them in front of a slit through which a beam of neutrons is allowed to pass after travelling down a flight tube from either a nuclear explosion or some sort of pulsed neutron source such as a chopper or linear accelerator target. In practice, however, it is far simpler to use mono-energetic neutrons from a nuclear reaction. (The term "monoenergetic" is not strict as there will always be a spread of neutron energy which depends on the solid angle of neutrons intercepted by the target, the thickness of the neutron target, the energy spread of the charged particle beam, etc.) Table 3 summarises the most useful of such reactions.

#### Table 3

## Mono-energetic neutron sources useful for fission yield measurements

Energy (KeV)	Most convenient reaction	Type of accelerator	Typical flux at O ⁰ ; 2 cm from neutron target: n/cm ² /sec	References to some papers using this source
30	${}^{7}_{Li(p,n)}{}^{7}_{Be}$	van de Graaff	$10^{7}$	12, 15
60	${}^{3}_{H(p,n)}{}^{3}_{He}$	""""	$10^{6}$	16
100-2,000	${}^{7}_{Li(p,n)}{}^{7}_{Be}$	van de Graaff or	$5 \times 10^{7}$	12, 15, 16, 57
*2,500-9,000	${}^{2}_{H(d,n)}{}^{4}_{He}$	Cockcroft-Walton	$5 \times 10^{7}$	16, 54, 55, 57
14,700	${}^{3}_{H(d,n)}{}^{4}_{He}$	Cockcroft-Walton	$5 \times 10^{8}$	15, 16, 56, 57

"The energy spread of the neutrons may become quite large above about 3 MeV for a variety of reasons and this must be carefully considered when planning experiments.

### (b) <u>Fission neutron sources with known spectra</u>

Much higher neutron fluxes can be obtained from some kind of fission source such as a critical assembly or full-scale reactor. It is, of course, necessary to know the neutron spectrum so that the effect on fission yields of changing it can be assessed. The most usual procedure is to make use of a low power assembly of some kind (flux :  $10^9 - 10^{11}$ ) and to change its core structure so that the neutron spectrum changes (51), but some information can be obtained by selecting different regions of an operating fast reactor whose spectra are well known (flux :  $10^{12} - 10^{14}$ )^(52,53). The difficulty with this last approach is that the neutron spectrum may change during the irradiation due to the operation of control rods or changes in position of other experiments. Finally it is possible to modify a neutron spectrum by the insertion of suitable obsorbers near to the substance being irradiated⁽⁵⁸⁾.

All these methods involve a neutron spectrum measurement at some stage of the experiment and this can be a very awkward process. It is to be hoped that it will be established that the yields of key nuclides required for reactor operation purposes are not affected by the changes in neutron spectra from one fast reactor to another so that one set of data can be used. This will not, of course, solve the problem of yield changes in wing and valley nuclides or of possible changes in the yields of nuclides affected by fine structure should these ever become of operational importance.

#### 3. EVALUATION OF FAST FISSION YIELDS

Evaluation procedures are dealt with in detail in paper 11a and this short section has been written only to point to the special problems confronting evaluators of fast yields as compared to thermal.

At the present time, and for reasons which are discussed under experimental methods (section 2), there are very few mass-spectrometric fast fission yields and not a great number of absolute yields either. This means that the evaluator cannot base his work only on accurate yields but must use all the available data, whose reliability is exceedingly variable. Furthermore, data on independent yields scarcely exist at all and so making allowance for them is difficult.

As if this was not enough, the evaluator of fast yields is also faced with a fundamental problem of fast fission - namely the effect of neutron energy and the fact that many experimenters have either not known or else ignored the shape of the neutron spectrum in which their measurements were made. The ideal situation would be to have a complete evaluation of all fast fission yields for every relevant neutron spectrum shape, but since this is utterly impossible now and probably always will be, all evaluators so far have chosen to ignore the problem and group all "fast" data together. The energy effect, if referred to at all, is dealt with

separately for the very few cases where any data exist. In fact, as we can see in the section of this paper on the yields themselves, these procedures have not resulted in huge discrepancies between evaluators, possibly because any energy effect is masked by the experimental errors.

The only possible recommendation for the future can be for experimenters to take more care in recording the experimental details of their neutron spectra and for evaluators so try and decide just how great the effect of neutron energy really is.

## 4. CHAIN YIELDS IN FAST FISSION

## 4.1 The available evaluations

If we exclude publications which are more than five years old, and also those in which the authors have not re-evaluated the original experimental data from first principles and thereby produced a new set of recommended yields, there are only four fast fission yield evaluations to consider; those of Crouch⁽¹⁾, Lammer and Eder⁽²⁾, Meek and Rider⁽³⁾ and von Gunten⁽⁵⁰⁾. I will now give a general description of the methods and principles adopted by these four sets of authors.

#### (a) Crouch

The UKAEA report by Crouch is the fast fission analogue of his earlier thermal yield evaluation paper⁽⁴⁾, which is included by Walker in paper 11a of this meeting. It contains recommended sets of data for  232 Th,  233 U,  235 U,  238 U,  237 Np and  239 Pu. Crouch is steadily working through a programme which involves the following steps:

- (1) Set up a system on the Harwell computer to store and provide rapid access to all experimental yield data (5,6).
- (II) Use the system to store a complete library of fission yields, and keep it up-to-date by continual search of the new literature.
- (III) Use this library and its associated computer programme to produce evaluations of cumulative fission yields as required. It is important to understand that the evaluations at this stage of the process are simply of experimental data, nuclide by nuclide; all the data for any particular fissioning species are normalised to reference yields which are obtained by assessment of all available absolute yields. Weighted means, simple means, and recommended values are given, but there is no other adjustment of the data,

e.g. by invoking the conservation laws; values for yields of chains for which there are no experimental data are only given if interpolating them is a simple matter, e.g. if there is no fine structure involved. Crouch includes all the experimental data as well as his evaluated figures. The two reports, refs. (1) and (4), are the first results of this type of procedure.

- (IV) Use the evaluation of stage (III) as the basis of a further evaluation in which physical laws such as conservation of mass, charge and energy, and calculated values of  $\bar{\nu}$  etc. are applied. This process provides a complete set of adjusted values for all mass chains, but it is important to understand that such procedures move the recommended values a further step away from the actual measured experimental results and, while giving a good overall picture, may yet be less accurate for any particular yield than the stage (III) process. Crouch has not yet completed this stage of his evaluation work.
- (b) Lammer and Eder

Their publication⁽²⁾ is mostly on thermal yields, but does include a recommended set of ²³²Th fast yields. As far as possible these authors have based their evaluations on mass spectrometric yields, but the paucity of such measurements for ²³²Th fast fission has meant that they have had to rely mainly on radiochemical ones, both of the "R-value" type (i.e. where uranium and thorium samples are irradiated simultaneously and count ratios obtained for particular nuclides), and of the relative yield type. The yield curve obtained by combining these measurements is finally normalised so that the heavy peak sums to 100%.

Lammer, in a contribution written for inclusion in this paper⁽⁷⁰⁾ but submitted too late for its data to be used in the comparisons between evaluations (para. 4.3), has commented in detail on the evaluation of  232 Th yields and has renormalised his data so as to include some new measurements. The effect of this is to change some his values, usually by less than 1%, but in some cases by considerably more. His original and new results are shown in Table 7 of Appendix 1.

He has also, in a second last minute contribution⁽⁷¹⁾ written a set of comments on  238 U fast fission yields. This includes a useful table of some of the more important yields, listing the available experimental data.

This table is reproduced in Appendix 3.

#### (c) Meek and Rider

Meek and Rider's evaluation⁽³⁾ includes recommended sets of fast fission yields for  232 Th,  235 U,  238 U and  239 Pu. Like Crouch, they have a complete computerised data base of all experimental measurements and they give these in their tables. All original data are renormalised to the estimated best reference value before being used in the evaluation.

For the evaluation itself they use a complete procedure whereby the chain yield is built up stage by stage for any mass by calculating the independent yields, averaging them with any existing weighted average experimental values, and then adding them together to give the calculated cumulative yield at that point in the chain. This value is then averaged with the weighted averaged experimental cumulative yield value (if any) so as to give the final cumulative yield at that point. This process continues along the chain until a stable member is reached. Finally, the cumulative yields for all stable members of the chain are added together to give the recommended chain yield. The complete set of chain yields for any fissioning nuclide is then normalised so as to total 100% for each mass peak.

# (d) von Gunten

von Gunten's method is not explained in detail in his paper, but it seems likely that he has obtained his recommended values of fission yields by averaging available experimental data and normalising to reference yields where necessary.

#### 4.2 The calculated yield curves of Sidebotham

When considering fast fission yield evaluations available today we must also take into account the yield tables produced by Sidebotham by calculation⁽⁷⁾. The purpose of these calculations is to provide sets of cumulative thermal and fast fission yields for fissioning species where the data are scanty or absent, and the author calculates such sets for fast fission of  232 Th,  233 U,  234 U,  237 U,  237 Np,  238 Np,  238 Pu,  240 Pu,  241 Pu,  242 Pu.  241 Am and  242 Cm, as well as for a number of thermal fission cases.

The computer program FISCAL⁽⁸⁾ is used. First of all, the mean fragment total kinetic energy, mean total prompt  $\gamma$ -decay energy, mean  $\overline{\nu}$  for heavy and light peaks and mean neutron energy are calculated by a variety

of empirical methods and normalised to known experimental values where possible. The mean fission fragment masses for light and heavy peaks are then calculated from these parameters by applying conservation of mass and energy during fission. The method of "mean mass shift" is now employed to obtain an unknown fission yield curve by taking a known curve and effectively shifting it along the mass axis in proportion to the difference in the mean masses of the two nuclides.

In the case of fast fission, the known curves used as the basis of the method are those for  235 U,  238 U and  239 Pu. The data for drawing these reference curves were taken from the older evaluations of Croall  $^{(9,10)}$ , supplemented by an earlier publication of Meek and Rider  $^{(11)}$  and are somewhat out-of-date.

#### 4.3 Comparison of the evaluations

The latest references quoted in the four evaluation papers are, Crouch (1972), Meek and Rider (1971), Lammer and Eder (1972) and von Gunten (1968), and we must assume that all published work up to these dates is included; in fact, the first two authors have exchanged their reference lists in the interest of completeness.

The main comparison in the evaluations must lie between Grouch and Meek and Rider, since they are the most complete sets. It is, therefore, important to understand that there is a difference in their methods of selection of the experimental data in that Grouch does not accept data unless it has, as a minimum requirement, been reported in the Transactions of the American Nuclear Society or in a final freely available report from a recognised labratory; Meek and Rider, on the other hand, include data taken from progress reports and similar more ephemeral documents.

Looking into the methods used by the evaluators to produce their recommended data sets, we can see that there is a difference in principle in that Crouch (and probably von Gunten) does not apply any constraints to the complete yield curves (although Crouch will do so when he produces his "Stage IV" evaluations), whereas the other two do. However, where the data are reasonably plentiful, as in the cases of  235 U and  239 Pu, we might expect any effect of this difference to be small, and so it seems reasonable to compare the resulting data sets directly. A set of tables has therefore been constructed (Appendix 1). These give the recommended values of the authors for yields > 0.001% and include a simple mean which has been

plotted in Figures 1 - 6 together with Sidebotham's values where relevant; these figures may therefore be considered to show the current best values for the fission yield curves for these fissioning species.

In order to show as graphically as possible the extent of the agreement between the evaluations, the percentage differences of the author's recommended values from their simple mean has been plotted in Figures 7 - 10. Sidebotham's values are also shown where relevant but they have NOT been included in the mean. These figures indicate that, if we only consider those data on the peaks of the mass yield curves (i.e. fission yields > 1%), the agreement between evaluations is not as bad as has often been supposed, being in round terms, from  $\pm 2\% - \pm 4\%$  for  $\begin{array}{c} 235 \\ \text{U} \text{ and } \end{array}$  $\pm$  8% -  $\pm$  10% for ²³⁸U, and  $\pm$  15% for ²³²Th. This does not, of course, necessarily mean that the experimental data are correct within these limits, but it is suggestive that they may be, particularly since the limits are broadly within the errors quoted by the evaluators for individual yields. It is worth remarking at this point that Crouch and Meek and Rider take considerable trouble to make estimates of the error attributed to every recommended yield, and that these errors do indeed seem reasonable in that they adequately cover the differences between the two evaluations. Lammer and Eder, for some reason, do not quote errors for their recommended values, except in a more general way.

On the whole, Sidebotham's calculated yields are somewhat further from the evaluator's means on average, but the discrepancies are not enormous, particularly on the peaks, and this seems to show that, in the absence of experimental data, his method can be used to provide at least a rough data set. It is clearly no substitute for careful experimental and evaluation work, however.

# 4.4 Discrepancies and shortcomings of the existing data

Undoubtedly the most important gap in the data for fast cumulative fission yields is the absence of any experimental information on  240 Pu and  241 Pu, and it is to be hoped that work at present in progress in various laboratories will rectify this before long. In the interim, the calculated data of Sidebotham can be used to show general trends.

The data are also very scanty for  233 U and  237 Np, and fairly discrepant for  232 Th and  238 U. In fact, only in the cases of  235 U and  239 Pu can we say that there is a fair amount of agreement, and even here we are

a long way from the sort of accuracy which has been achieved for the corresponding thermal fission yields.

Table 4 contains my rough estimates, based on the degree of unanimity achieved by the evaluators and on the errors which they ascribe to the experimental data, of the level of accuracy we have attained at the present time.

## Table 4

level of accuracy	(10	) of	fast	fission	yields	at	t he	present	time
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Figgioning	Average accuracy (10) of fast yields : $\%$						
Nuclide	Light peak (yields > 1%)	Heavy peak (yields > 1%)	Wings and Valley				
232 _{Th}	± 15	± 10	± 30				
233 _U	± 15	± 15	± 20				
235 _U	± 7	± 4	± 15				
238 _U	± 10	± 10	± 20				
237 _{Np}	± 15	± 15	insufficient data				
239 _{Pu}	± 7	± 4	± 15				

In general, it is for burn-up measurements that the most accurate yields are needed. Table 5 contains my estimates of the accuracy at present achieved for yields used for this purpose for the three most important fissioning species. I have based this assessment on the quoted errors of the evaluators.

#### Table 5

Burn-up	Estimated accuracy (10) achieved now: $\%$					
Nuclide	235 _U	238 _U	239 Pu			
95zr./Nb	3	7	3			
¹³⁷ Cs	6	10	5			
140 _{Ba/La}	2	3	2			
143 _{Nd}	3	8	3			
145 _{Nd}	3	8	3			
146 _{Nd}	3	8	3			
148 _{Nd}	3	8	3			
150 _{Nd}	4	10	3			
1		I	1			

# Degree of accuracy (10) of fast fission yields used for burn-up at the present time

#### 4.5 Recommendations for future work

For fast reactor purposes really accurate fission yields are needed only for nuclides for monitoring, largely those mentioned in Table 5. All these yields need to be improved for  238 U but, with the possible exception of  137 Cs, the present data are probably just about good enough for  235 U and  239 Pu. (The consensus of opinion is that 2 - 3%, 10, is the accuracy required for monitor nuclides.) For all other fuel nuclides, particularly  232 Th,  233 U,  240 Pu and  241 Pu, the monitor data are nowhere near adequate, and therefore more measurements must be made. Note that fission yields are also needed when fast fission threshold monitors are used for integrated neutron dose or fluence measurements. This extends the data requirement for Table 5 nuclides to such substances as  231 Pa and  236 U.

As far as the other yields are concerned, high accuracy is not required for the strictly utilitarian purpose of reactor design and operations, although it is desirable to have better data for the use of those who are trying to understand fission from a fundamental standpoint, in itself a study which must ultimately benefit the reactor designer. Good overall data also make. evaluation work easier and this too cannot help but be an advantage to reactor projects in the long run. As a first priority, future work on the non-monitor nuclides should therefore concentrate on  240 Pu and  241 Pu, followed by  232 Th,  233 U and  238 U.

Information sent to me for this meeting suggests that yield measurements in progress in the UK and the USA (Crouch, Davies, Maeck, Larsen) should go some way towards satisfying the above requirements, at least for  235 U,  238 U,  239 Pu,  240 Pu and  241 Pu, although it does not seem likely that they will do so completely. On the evaluation side, we may expect further outputs from the computer-based data sets of Crouch and Meek and Rider and, it is to be hoped, from Deviliers also. It seems imperative that the three authors of the computer-based evaluations try and co-operate to the largest extent possible. It also seems important that the new data reported in section 4.6 are rapidly included in evaluations, since they considerably increase the information on  232 Th,  255 U,  238 U and  239 Pu.

# 4.6 New data

The following is a short summary of fast yield data which have not yet been included in any of the current evaluations. These data are a valuable increment to present information and should be evaluated as soon as possible.

# (a) <u>Matthews and Tomlinson</u> (66)

This is a considerable addition to the fast yield information on ²³⁸U, since it contains mass-spectrometric yields of Xe, Cs, Ba, Ce, Nd and Sm, totally 20 mass chains. The yields are normalised to each other and then made semi-absolute by normalising the heavy peak to 100%. Irradiations were carried out inside a Cd sheath in a swimming pool reactor. No detailed information about the fast neutron spectrum is given and any possible energy effect is ignored.

# (b) Larsen et al. (51)

This paper, in which the authors report absolute yields of  95 Zr,  97 Zr,  103 Ru,  131 I and  140 Ba measured in fast (and thermal) fission of  235 U and  239 Pu, and fast fission of  238 U, is an amplification of an earlier one  ${}^{(18)}$ . The work was done mainly to investigate the effect of neutron energy and is also discussed in section 6.3.

(c) Blachot and Chauvin⁽⁶⁵⁾

Relative yields of 22 mass chains in the reactor neutron fission of  $^{\rm 232}_{\rm Th.}$ 

(d) <u>Maeck</u>⁽⁶⁷⁾

Absolute yield measurements have just been completed for fission of  235 U in a neutron spectrum characteristic of a liquid metal fast breeder reactor. The data comprise ~ 95% of the heavy mass peak yields and ~ 75% of the light and are summarised in Figure 11, supplied by the author. He also has preliminary data for  238 U and  239 Pu. Presumably this work is a continuation of that reported in reference (50).

(e) Grütter and von Gunten⁽⁶⁸⁾

These authors have determined 20 yields in the mass ranges 91 - 105 and 128 - 151 for fission of  235 U in the Proteus reactor whose neutron spectrum is similar to that of a helium cooled reactor. Their method was to use accurate  $\gamma$ -spectrometry and to make the measurements relative to  235 U samples irradiated in a thermal neutron spectrum.

5. CHAIN YIELDS IN 14 MeV FISSION

#### 5.1 The available evaluations

14 MeV fission yields are of negligible importance in fission reactors and will therefore be dealt with only in outline in this paper. They have, of course, considerable interest for those concerned with some thermonuclear work, and a study of them is relevant for those engaged in fundamental fission research. The evaluations currently recommended are those of von Gunten⁽⁵⁹⁾, Meek and Rider⁽³⁾, Crouch⁽¹⁾ and Daróczy, Raics and Nagy⁽⁶⁰⁾. The main aspects of these papers will now be very briefly mentioned.

# (a) von Gunten

This is the oldest of the four and contains recommended sets of yields for  232 Th,  235 U and  238 U. It is not clear from the text exactly what procedure was used to obtain the recommended values, but it seem probable that they are simple means of all the data from the quoted authors. No suggested errors are given for the recommended values.

## (b) Meek and Rider, and Crouch

Meek and Rider give recommended data sets for  $\begin{array}{c} 235\\ \text{U} \text{ and } \end{array}$  U, and Crouch for  $\begin{array}{c} 231\\ \text{Pa}, \end{array}$  Th, U, U, Np, U, Pu and  $\begin{array}{c} 238\\ \text{V}, \end{array}$  Pu.

The methods used by these authors have already been discussed in section 4, on fast fission yields.

# (c) Daróczy, Raics and Nagy

This evaluation was carried out especially for inclusion in this paper, but it is to be hoped that the authors will publish it in full elsewhere since it is impossible to include more than a summary here; it is for 238 U only.

The authors first took all available absolute yields for ⁹⁹Mo and ¹⁴⁰Ba (including absolute ratios of the two yields) and prepared weighted averages of them. All other yields were then normalised to these values (except in a few cases where this was not possible) and simple and weighted average yields were calculated for every nuclide for which experimental data exists. Like Crouch, these authors have not applied general constraints, such as normalisation of peaks to 100%, to their results. Carefully estimated errors are quoted for all yields.

# 5.2 Comparison of the evaluations

Although the methods used by the four evaluators are slightly different, the quality of the data is such that differences between them cannot be separated from the errors. For presentation purposes, therefore, it seems simplest to display curves of the means of the evaluated yields, and these are shown in Figures 12 to 17. The data themselves are given in Appendix 2, Tables 1 to 6. The data of Daróczy, Raics and Nagy have also been shown separately in a curve supplied by them (Figure 18) since their evaluation is the latest and includes some data not used by the other authors.

# 5.3 Discrepancies and shortcomings of the existing data

None of the existing data sets is comparable in accuracy with those for the thermal yields of the fuel nuclides, and those for  237 Np and  239 Pu are sparse indeed. However, these data are of little importance to the operation of fission reactors and therefore, as far as the terms of reference of this paper are concerned, are probably adequate. Table 6 gives my estimates of the general level of accuracy of these yields at the present time.

#### Table 6

Fissioning	Average accuracy (10) of 14 MeV yields ; $\%$					
Nuclide	Light peak Heavy peak (yields 1%) (yields 1%)		Wings and Valley			
232 _{Th}	± 15	± 15	± 30			
233 _U	± 15	± 15	± 30			
235 _U	± 10	± 10	± 25			
238 _U	± 10	± 10	± 20			
237 _{Np}	± 20	± 20	insufficient data			
²³⁹ Pu	± 15	± 15	insufficient data			

## Level of accuracy (10) of 14 MeV fission yields at the present time

#### 5.4 New data

Daróczy et al. have included data of their own in their evaluation which is to be published in two papers:-

(a) Daróczy, Nagy, Kover, Raics and Csikai⁽⁶²⁾

5 absolute chain yields in  238 U 14 MeV fission, together with a measurement of the fission cross-section.

(b) Daróczy, Germán, Raics, Nagy and Csikai⁽⁶³⁾

17 absolute chain yields in  238  U 15 MeV fission.

In addition to these, some new data have appeared, or will appear, in the literature which are not yet included in any of the evaluations:-

(c) <u>Nethaway and Mendoza</u>⁽⁶¹⁾

24 chain yields for  234 U 14 MeV fission made as relative yields and then normalised to 200%. 30 absolute chain yields for  236 U 14 MeV fission.

- (d) <u>Bocquet, Brissot, Crancon and Moussa⁽⁶⁴⁾</u>
   Kr and Xe yields for 14 MeV fission of ²³³U, ²³⁵U, ²³⁸U and ²³²Th.
- (e) <u>Blachot, Carraz, Cavallini, Chauvin, Ferrieu and Moussa⁽⁵⁶⁾</u>
   Absolute yields for 28 mass chains in 14 MeV fission of ²³⁸U.

# (f) Blachot and Chauvin⁽⁶⁵⁾

Absolute yields for 28 mass chains in 14 MeV fission of  $\frac{232}{Th}$ .

(g) Mandler, Reed and Moler⁽⁵⁷⁾

Relative yields for 20 mass chains for  235  U and  238  U fission at various energies, including 14 MeV.

These papers provide a considerable amount of new data in 14 MeV fission, and it is desirable that they should be included in new evaluations before long. However, a preliminary cursory examination suggests that, while they will somewhat improve the quality of the existing data, they do not significantly alter the conclusions given in section 5.3 above, particularly as no new measurements are reported on 237 Np or 239 Pu.

# 6. THE EFFECT OF NEUTRON ENERGY ON FISSION YIELDS

#### 6.1 General discussion of the problem

In general terms there must, of course, be a substantial change in fission yield values if there is a substantial change in the energy of the neutrons causing the fission. The dominance of the asymmetric modes in fission of heavy nuclides is a low energy phenomenon caused by the nuclear shell structure, which gradually washes out as excitation increases, resulting in the fission becoming more and more symmetric. However, at the low excitation energies produced by reactor neutrons this effect is quite small and can be readily observed only in the rare fission modes seen in the valley and on the wings of the yield curve. There must be some compensating effect on the peaks, but the question we are really trying to resolve is whether or not this is negligible for the practical purposes of nuclear reactor designers and operators.

In fast reactors the neutrons cover an energy band which varies from reactor to reactor, but is substantially between 1 KeV and 10 MeV, with a mean which is usually between 100 KeV and 1 MeV, and so, at least for reactor purposes, the possible effects we are looking for are those caused by a mean excitation of at most 1 MeV above thermal values. Thus the most important energy range for comparative measurements is that from about 10 KeV to 2 MeV.

The experimental data, which are exceeding sparse, result from two different types of measurement, integral in which the sample is irradiated in reactor neutron spectra which differ in shape, and differential in which mono-energetic neutrons are used. Note that the vast majority of integral measurements of fast fission yields have been made in an unspecified spectrum and so do not add to the available data on the effect of energy, except to provide a sort of base-line set of typical "fast" fission yields.

#### 6.2 Mono-energetic neutron experiments

The available data have been well tabulated in ref. (1) and there seems little point in repeating these tables here. Only two papers have appeared since ref. (1) was prepared, by Cuninghame, Goodall and Willis⁽¹²⁾, and by Mandler. Reed and Moler (57). I have shown the main results from ref. (12) in Table 7 in order to supplement ref. (1). Table 8 shows the results from ref. (57) in the form given by the authors, i.e. as yield ratios relative to the mass 140 chain, with the 15 MeV ratios made equal to their values in ref. (69). Unfortunately, insufficient information is in the paper to allow me to convert these ratios to individual fission yields and so I have been unable to compare them with others shown in Figures 17 - 21. This is a pity because there is a considerable amount of valuable data here and I hope that the authors will carry out the conversion themselves before long. Some mono-energetic experiments have measured fission yields at only one energy above thermal (often an energy of several MeV) and such information by itself is little help when it comes to trying to decide whether or not the yields change over the range of neutron energies to be expected in a fast reactor. Furthermore, about half of the results are for yields of muclides in the valley or on the wings of the mass yield curve and so are barely relevant to practical reactor problems, while a large proportion of the results with peak nuclides give little information on how the yields fare as the neutron energy changes over the critical 10 KeV - 2 MeV region.

The present position is summarised in Table 9; 14 MeV fission is not included in this table since it is dealt with separately in section 5 of this paper but far more data have been collected at this energy, undoubtedly because it is the only one at which adequate neutron fluxes can easily be generated. Where the table shows data to be available in the range 2 - 14 MeV, they are mainly at 3 and 8 MeV with occasional measurements at other energies. Data in the 10 KeV - 2 MeV range for  238 U are at two points only, 1.5 and 2 MeV. Thus it is only in  235 U and  239 Pu fission in the range 10 KeV - 2 MeV that there are data at a reasonable number of energies, and then only for a few nuclides; for peak nuclides there are only the data of ref. (12) and (57). Figures 19 - 23 and 25 show all the data for  235 U and  239 Pu

Nuclide	N <b>eutron en</b> ergy KeV	Fission yield
99	ан на настраниятеля (18262); запалениения, талиналенно (18252) 	r 70 ± 0 70
NO.	130	5.70 2 0.39
	700	5 60 + 0 32
	900	$5.78 \pm 0.32$
	1300	5.55 + 0.33
	1700	5.55 ± 0.37
TIL	t 3/)	0.015 + 0.002
Ag	300	$0.013 \pm 0.002$
	700	0.024 1 0.003
	900	$0.024 \pm 0.005$
	1300	0.044 ± 0.002
	1700	0.053 ± 0.004
140 ₈₀	1 7/7	6 10 + 0 Au
2342	300	6.79 + 0.35
	700	$6.08 \pm 0.36$
	\$60	$6.01 \pm 0.35$
	1300	5.97 + 0.36
	1700	5.75 ± 0.32
147 _{N/I}	1.30	2.35 + 0.14
1.112	300	$2.61 \pm 0.16$
	700	2.51 ± 0.18
	\$00	2.46 ± 0.14
	1 300	2.56 ± 0.17
	1700	2.42 ± 0.13
153 _{Sm}	1.30)	0,132 + 0.013
	300	0.152 ± 0.009
	700	0,154 ± 0.010
	900	$0.159 \pm 0.014$
	1300	0.176 ± 0.013
	1.500	1 0 170 1 0 010

.

 $\frac{\text{Table 7}}{\text{Absolute fission yields of }}$ 

Table	-8
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Ratios of fission yields relative to the yields for the mass 140 chain (Ref. (57))

Mass			U	235			U-238				
Chain	1 MeV	2 MeV	4 MeV	6 MeV	9 MeV	15 MeV	2 MeV	4 MeV	6 MeV	9 MeV	15 MeV
91	1.03	1.10	1.05	1.04	1.06	1.06	0.62	0.66	0.64	0.65	0.78
	±.07	±.05	±.06	±.08	±.06	±.09	±.02	±.03	±.03	±.03	±.04
92	0.98	0.95	1.15	1.09	1.08	1.08	0.73	0.84	0.80	0.81	0.87
	±.06	±.11	±.10	±.09	±.07	±.11	±.06	±.09	±.09	±.13	±.07
93	0.97	1.11	1.12	1.07	1.04	1.09	0.71	0.84	0.78	0.78	0.96
	±.03	±.05	±.06	±.06	±.04	±.11	±.02	±.04	±.04	±.04	±.07
95	1.11	1.16	1.20	1.13	1.14	1.09	0.91	1.00	0.95	0.97	1.13
	±.05	±.04	±.06	±.06	±.05	±.09	±.02	±.04	±.03	±.03	±.03
97	1.13	1.19	1.24	1.22	1.22	1.22	1.03	1.14	1.09	1.08	1.24
	±.04	±.05	±.07	±.06	±.07	±.11	±.03	±.07	±.05	±.06	±.07
99	1.11	1.19	1.21	1.18	1.18	1.12	1.08	1.16	1.08	1.03	1.23
	±.03	±.04	±.06	±.05	±.05	±.10	1.04	±.04	±.04	±.05	±.04
103	0.64	0.67	0.75	0.74	0.67	0.76	1.00	0.98	0.91	0.94	0.97
	±.03	±.02	±.05	±.05	±.03	±.06	±.04	±.06	±.04	±.04	±.04
105	0.19	0.21	0.26	0.29	0.23	0.37	0.76	0.75	0.70	0.72	0.74
	±.01	±.01	±.02	±.02	±.01	±.04	±.03	±.05	±.03	±.03	±.03
112	0.014	0.009	0.034	0.056	0.038	0.24	0.0080	0,014	0.023	0.026	0.150
	±.001	±.002	±.005	±.002	±.004	±.02	±.0007	<u>+</u> 002	±.001	±.003	±.006
127	0.051	0.076	0.166	0.240	0.140	0.50	0.019	0.040	0.079	0.073	0.37
	±.002	±.004	±.009	±.009	±.005	±.02	±.002	±.002	±.003	±.003	±.02
129	0.22	0.20	0.29	0.37	0.28	0.71	0.17	0.13	0.24	0.15	0.64
	±.02	±.02	±.02	±.02	±.02	±.05	±.03	±.03	±.03	±.04	±.04
131	0.68	0.75	0.87	0.89	0.78	0.93	0.64	0.74	0.78	0.74	1.05
	±.04	±.04	±.06	±.06	±.05	±.09	±.03	±.05	±.05	±.04	±.05
132	1.01	0.99	1.07	1.21	1.04	1.09	0.90	0.89	0.96	0.86	1.02
	±.05	±.04	±.06	<u>+</u> .06	±.05	±.09	±.03	±.05	±.04	±.04	±.03
133	1.13	1.24	1.25	1.30	1.16	1.18	1.36	1.39	1.36	1.39	1.44
	±.09	±.08	±.10	±.09	±.11	±.13	±.09	±.13	±.09	±.11	±.10
135	1.44	1.65	1.57	1.46	1.45	1.24	1.13	1.17	1.09	1.12	1.18
	±.07	±.06	±.11	±.07	±.05	±.08	±.04	±.07	±.05	±.04	±.04
137	1.01	1.01	0.95	1.00	0.92	1.10 [.]	1.30	1.04	1.16	0.96	1.16
	±.06	±.07	±.11	±.06	±.05	±.07	±.07	±.08	±.06	±.10	±.08
143	0.97	1.01	0.95	0.89	0.96	0.85	0.67	0.73	0.71	0.69	0.78
	±.09	±.06	±.08	±.07	±.08	±.11	±.04	±.06	±.05	±.03	±.04
144	0.56	0.52	0.73	0.51	0.51	0.72	0.64	0.54	0.63	0.65	0.72
	±.08	±.09	±.25	±.13	±.09	±.09	±.07	±.07	±.05	±.08	±.07
147	0.48	0.49	0.48	0.52	0.50	0.44	0.39	0.43	0.41	0.42	0.43
	±.02	±.02	±.04	±.03	±.03	±.03	±.01	±.01	±.01	±.01	±.01
151	0.041	0.043	0.055	0.C54	0.046	0.045	0.139	0.168	0.162	0.136	0.187
	±.002	±.002	±.003	±.004	±.003	±.010	±.006	±.010	±.010	±.005	±.007

# Table 9

# Available mono-energetic neutron data relating to the effect of energy on fission yields

Fissile Nuclide	Part of mass yield curve covered	Measurements in range 10 KeV-2 MeV	Measurements in range 2 MeV-14 MeV (excluding 14 MeV)
232 _{Th}	Light wing (LW) Light peak (LP) Valley (V) Heavy peak (HP) Heavy wing (HW)	No No No No	Yes Yes Yes Yes Yes
233 _U	LW	No	No
	LP	No	No
	V	No	No
	HP	No	No
	HW	No	No
235 _U	LW	No	No
	LP	Yes: $97_{Zr}$ , $99_{Mo}$	Yes
	V	Yes: $111_{Ag}$ , $113_{Ag}$ , $115_{Cd}$	Yes
	HP	Yes: $140_{Ba}$ , $143_{Ce}$ , $147_{Nd}$	Yes
	HW	Yes: $153_{Sm}$	Yes
238 _U	LW	Yes: ⁷⁷ As	Yes
	LP	No	Yes
	V	Yes: ¹¹¹ Ag, ¹¹⁵ Cd	Yes
	HP	^{No}	Yes
	HW	No	Yes
²³⁹ Pu	LW LP V HP HW	No No Yes: ¹¹¹ Ag, ¹¹³ Ag (plus measurements in individual resonances in range 0.06-0.36 eV) No No	No No No No No

for nuclides where a fair number of yields at different energies have been reported. Note that some points from measurements made in reactors have been included by plotting evaluated fission yields at an energy of 0.6 MeV, assumed to be a reasonable mean fission energy in these cases; on the whole these reactor measurements are in agreement with the mono-energetic points except for the case of 147Nd. It seems possible that the data of ref. (12), which are absolute yields, may for this nuclide harbour some unsuspected systematic error. This possibility does not, however, invalidate the main conclusion (b) below.

We can summarise the results of the mono-energetic measurements as follows:-

- (a) Valley and wing yields rise with energy as expected.
- (b) More tentatively, yields on the peaks are constant over the range of neutron spectra to be expected in fast reactors within about  $\pm 4\%$ , but start to fall off above ~ 2 MeV.
- (c) There is no available information on changes in yields of nuclides affected by fine structure.

# 6.3 Integral experiments

Even less work has been reported on integral measurements, but what there is helps to confirm the tentative conclusion (b) in section 6.2 above. Larsen et al. ⁽¹⁸⁾ carried out experiments in a fast reactor with a variable core configuration (ZPR-3) from which they concluded that the yields of ²³⁵U and ²³⁹Pu burn-up and dosimetry monitors vary < 1% within an energy range from the core to the nickel reflector, i.e. between median fission energies 446 KeV - 0.37 KeV. These authors have now extended their earlier work⁽⁵¹⁾. They have measured the yields of ⁹⁵Zr, ⁹⁷Zr, ¹⁰³Ru, ¹³¹I and ¹⁴⁰Ba in fast fission of ²³⁵U and ²³⁹Pu by accurate  $\gamma$ -spectrometry, the number of fissions being determined by fission track detectors. They now find that over the neutron energy range thermal - 500 KeV the relative changes of the ²³⁵U and ²³⁵U fission products are all less than 1%. In the case of ²³⁵U, however, the yields of ¹⁰³Ru and ¹³¹I increase with the logarithm of the neutron energy.

Another piece of information has come from B. F. Rider who has sent some advance results of calculations which will appear in the 1974 edition of the Meek and Rider fission yield evaluation. He finds that  235 U fast yields in the valley increase by a factor of 2.8 over thermal values, and therefore calculates that peak yields must fall by a factor of 0.97 of thermal to compensate; for  239  Pu the corresponding figures are an increase in the valley by 1.8 and a fall on the peaks of 0.98 - 0.99. Taking this in conjunction with the observed increase in the valley over the crucial fast reactor energy range as seen in the mono-energetic experiments, we can see that any corresponding reduction in peak yields will be at most 2 - 3%.

### 6.4 Gaps in the data and recommendations for future work

As far as it goes, the evidence presented above confirms what one would intuitively expect, i.e. for the peak nuclides the variation of fission yields over the range of neutron energies important to a fast reactor is very small and can probably be ignored. However, the actual amount of experimental information is minimal, the only reported data being for  235 U and  239 Pu; it is certainly desirable that confirmatory experiments are carried out for other fuel nuclides.

For wing and valley nuclides the evidence is, again as expected, that the fission yields rise with neutron energy. It seems essential to make many more measurements of these yields because the Nd isotopes are used for monitoring. ¹⁵⁰ Nd, at least, may well change by an amount large enough to matter and the existing data are very skimpy and again restricted to ²³⁵ U and ²³⁹ Pu.

Finally there is the question of nuclides in the fine structure region around mass 135. There are no precise experimental data showing how yields in this region change with neutron energy, but it is certainly possible that they may do so more rapidly than the average peak nuclide. Since any such change might affect the yield of 137Cs, new data are urgently needed here also.

#### 7. CHARGE DISTRIBUTION IN FAST AND 14 MeV FISSION

#### 7.1 General discussion

Experimental information on the way in which the protons from the fissioning nucleus are distributed relative to all the nucleons when fission occurs is obtained by measuring the fission yields of individual fragments formed in the primary fission act ("independent fission yields"). The measurements must be made before earlier members of the mass chain concerned have decayed into the nuclide being measured, or else corrections must be made for such decay. This makes the experiments difficult to carry out except in a rather limited number of cases, and so the tally of values available in the literature is small - particularly so in the case of fission above thermal energies.

## 7.2 Measured independent yields

All the available experimental independent yields for fission by neutrons above thermal energies ar i including 14 MeV are given in Table 10. They are shown divided into "fast and "14 MeV" groupings, the former including all measurements below 14 MeV. The table gives the "fractional chain yield" (i.e. the fraction of the total chain yield which is formed directly as the nuclide concerned) rather than the independent yield itself since presenting the yields in this way does not involve using the actual chain yield value.

It is difficult to know how best to attempt any correlation of such a very few results, frequently having large experimental errors, and coming from a number of very diverse types of fission. Ideally we would like to be able to analyse the independent yields of all the members of a large number of mass chains all for the same type of fission but such data are simply not available. In the event, it seems best simply to plot all the results as a function of  $(Z-Z_p)$  where  $Z_p$ , the most probable charge for a particular mass chain, is that calculated by Crouch⁽²⁹⁾ as extrapolated by Meek and Rider⁽³⁾ or by myself (see section 7.3 below).

The resulting correlation is given in Figure 24. Note that some of the points (marked on the error bar with an arrow  $\wedge$ ) are for single isomers of a pair and are therefore only lower limits.

## 7.3 Calculated independent yield

For many purposes, including some methods of evaluating chain yields, we must have independent yields for all or most nuclides formed in fission. It is quite clear that we are nowhere near having measured values for all these yields, nor are we likely to have them in the foreseeable future. Calculation of yields entirely from fission theory is also something not likely to be achieved for a long time to come, and so the best possible compromise seems to be to calculate them by imposing some reasonable theoretical constraints and making use of the available experimental data to provide such constants as are needed for the calculations. Actual methods of calculation are outside the terms of reference of this paper, since it is mainly a correlation of data, but a brief summary is given below.

# Table 10

System	Nuclide	Fractional yield	z _p	Reference
²³² Th fast	^{131m} Te ¹³³ I ¹³⁴ I ¹³⁵ J ¹³⁶ Cs	$(9.6 \pm 3.1) \times 10^{-3}$ (4.8 ± 0.8) × 10 ⁻² (2.9 ± 0.5) × 10 ⁻² (1.41 ± 0.45) × 10 ⁻¹ (2.40 ± 0.77) × 10 ⁻⁵	50.22 51.00 51.42 51.81 52.18	19 20 20 20 20 21
²³² Th 14 MeV	$95m+g_{Nb}$ $96_{Nb}$ $99m_{Tc}$ $106_{Rh}$ $112_{Ag}$ $115m_{In}$ $117m+g_{In}$ $124m+g_{Sb}$ $126m+g_{Sb}$ $130m+g_{I}$ $132_{I}$ $133_{I}$ $134m+g_{I}$ $134m+g_{I}$ $134mc_{S}$ $135m_{Cs}$ 170	$(3.4 \pm 0.8) \times 10^{-2}$ $(1.7 \pm 0.4) \times 10^{-2}$ $(1.8 \pm 0.4) \times 10^{-3}$ $(7.2 \pm 1.4) \times 10^{-3}$ $(1.2 \pm 0.3) \times 10^{-2}$ $(1.5 \pm 0.3) \times 10^{-2}$ $(1.4 \pm 0.4) \times 10^{-2}$ $(1.4 \pm 0.4) \times 10^{-2}$ $(1.0 \pm 0.3) \times 10^{-1}$ $(1.0 \pm 0.3) \times 10^{-1}$ $(1.5 \pm 0.4) \times 10^{-2}$ $(3.6 \pm 0.7) \times 10^{-2}$ $(1.2 \pm 0.3) \times 10^{-1}$ $(8.0 \pm 2.0) \times 10^{-2}$ $(9.5 \pm 2.5) \times 10^{-2}$ $(1.52 \pm 0.5) \times 10^{-3}$ $(2.4 \pm 0.5) \times 10^{-2}$	38.64 38.97 39.94 42.34 44.96 46.43 47.27 49.23 49.83 50.57 51.22 51.65 52.07 52.17 52.46	22 22 22 22 22 23 24 24 24 23 23 23 23 20 23 20 23 20 23 20 22 22
²³⁵ U 14 MeV	131m _{Te} 131g _{Te} 1311 1321 133g _{Te} 133g _{Te} 133 ₁ 133 _{xe}	$(2.0 \pm 0.3) \times 10^{-1}$ $(3.5 \pm 0.56) \times 10^{-1}$ $(2.98 \pm 0.95) \times 10^{-1}$ $(8.00 \pm 2.56) \times 10^{-2}$ $(1.61 \pm 0.13) \times 10^{-1}$ $(4.08 \pm 1.31) \times 10^{-1}$ $(4.08 \pm 1.31) \times 10^{-1}$ $(2.2 \pm 0.7) \times 10^{-2}$	52.73 50.80 50.80 50.80 51.79 52.22 52.22 52.22	24 25 25 25 25 25 25 25 25 26

# Fractional chain yields in fast and 14 MeV fission

System	Nuclide	Fractional yield	Z _p	Reference
²³⁵ U 14 MeV	¹³⁴ I	$(4.04 \pm 0.65) \times 10^{-1}$	52.63	25
(cont'd)	¹³⁴ Xe	$(2.6 \pm 0.1) \times 10^{-1}$	53.04	26
²³⁸ U fast	131m _{Te}	$(3.63 \pm 1.16) \times 10^{-2}$	50.46	19
	136 _{Cs}	$(2.31 \pm 0.74) \times 10^{-5}$	52.12	21
²³⁸ U 14 MeV	$92_{Y}$ 124 _{Sb} 126 _{Sb} 133 _{Xe} 135 _{Xe} 136 _{Cs} 140 _{La}	$(7.28 \pm 2.33) \times 10^{-3}$ $(2.53 \pm 0.81) \times 10^{-2}$ $(7.88 \pm 5.04) \times 10^{-3}$ $(3.0 \pm 1.3) \times 10^{-3}$ $(5.6 \pm 3.7) \times 10^{-2}$ $(9.02 \pm 2.89) \times 10^{-3}$ $(7.48 \pm 0.30) \times 10^{-3}$	36.61 49.07 49.61 51.63 52.41 52.77 54.28	27 24 24 26 26 26 24 27
²³⁹ Pu fast	135m _{Xe}	$(1.46 \pm 0.93) \times 10^{-2}$	52,61	28
²³⁹ Pu 14 MeV	¹³³ Xe	$(1.2 \pm 0.01) \times 10^{-1}$	52 <b>.</b> 43	26
	¹³⁵ Xe	$(4.6 \pm 0.3) \times 10^{-1}$	53 <b>.</b> 16	26

Table 10 (cont'd)

.

The most complete sets of calculated independent yields will be found in Meek and Rider⁽³⁾ and Crouch⁽²⁹⁾, in which the authors have used the method of Wahl⁽³⁰⁾ as a basis. In this method experimental independent yields are fitted to gaussian distributions for particular mass chains where there are sufficient of them to do this. The gaussian width parameter, C, can then be measured and averaged for all the different chains; a value of 0.62 (based on thermal fission yields) was deduced by Wahl and used by Crouch (except where direct experimental evidence suggested some other value), while Meek and Rider used a later, improved, Wahl value⁽³¹⁾ of 0.56. The other parameter required in the calculation, Z_p is evaluated for a particular mass chain from the equation

$$Z_p = A\left(\frac{Z_F}{A_F}\right) + (correction factor)$$

where A is the mass number for the primary fission fragment (i.e. before prompt neutron emission) and  $Z_F$  and  $A_F$  are the atomic and mass numbers of the fissioning nucleus. To evaluate A we must have a curve showing the dependence of prompt neutron emission on fission fragment mass. The "correction factor" is estimated from known independent yields.

## 7.4 Errors

The errors on independent yield measurements reported by evaluators and experimenters are of the order of  $\pm$  20% on average, with a very few claimed to be as low as  $\pm$  10% and a few given as high as  $\pm$  60%. Because so few of the yields have been independently measured by different authors it is difficult to assess whether such errors are realistic or not; in fact the only case of such duplication is in the yields of ¹³³I and ¹³⁴I for ²³²Th 14 MeV fission, where the results are in agreement within the reported errors.

The crude correlation given in Figure 24 gives some indication of the reliability of the quoted errors because it is probably reasonable to expect that the points for any particular fissioning system should fall somewhere near to a gaussian fit. We can see that, while there is a general trend in that direction, the fit is certainly not within the experimental errors. It appears, therefore, that the best advice that can be given at this time is to regard the quoted experimental errors as being random only, and to assume that there may always be a considerable systematic error not allowed for.

If independent yields are needed, it is probably best to use calculated ones which, we hope, may be improved along the lines suggested in section 7.5 below before too long. The authors of the existing calculations have not given any errors; however, since the fractional yields for each mass chain must total to unity it seems clear that errors on yields close to  $Z_p$ cannot be very large - say  $\pm$  10% at most, provided we are correct in assuming that they really do fall on a gaussian curve. Errors in the yields of members of the chain further from  $Z_p$  might be more than this, since they are very dependent on the value of  $\sigma$  used but, being much smaller, these yields are correspondingly less important. It is probably safe to assume that the calculated independent yields are at least as accurate as the existing experimental ones.

#### 7.5 Gaps in the data and recommendations for future work

Experimental measurements in this field being so scarce, it is more a question of data in the gaps rather than gaps in the data. To suggest, however, that all the gaps ought to be filled immediately is quite unrealistic because of the immense practical difficulties in this type of work, and there is no doubt that the most fruitful course for the immediate future is to make independent yield measurements which will help to improve the necessary input parameters for the calculations and to refine the calculations themselves and extend them to fissioning systems of importance to users which are not yet covered. At present the only fast fission systems included in Crouch's publication are  232 Th,  238 U and  240 Pu, while Meek and Rider have only fast fission of  232 Th,  235 U,  238 U and  239 Pu and 14 MeV fission of  235 U and  238 U.

In order to be able to refine these calculations and extend them to other systems it is essential to have more information concerning the functions  $\sigma$  and  $z_p$ . In practical terms this means that experimenters should try to measure as many independent yields for any one mass chain as possible, and repeat the measurements for as may different mass chains as possible. It also means that far more data should be collected on prompt neutron emission as a function of fragment mass for the different fissioning systems. It is quite clear that a large amount of effort is needed to raise the level of fast neutron independent yield data even to that at present existing for thermal fission.

### 8. TERNARY FISSION

# 8.1 Definition

In ternary fission the fissioning nucleus breaks up into three, instead of into two particles. It is usual to include in this kind of fission only those cases where the third particle is charged, and this practice is followed here. Ternary fission events are rare, of the order of one per several hundred fissions, but a wide variety of particles may be emitted, from protons to fragments in the mass range 20 - 60. The subject has been reviewed twice recently (32, 33).

# 8.2 The data

The third particles whose presence has been reported in ternary fission are ¹H, ²H, ³H, ³He, ⁴He, ⁶He, ⁸He, Li, Be and heavier particles. As a typical example of the yields of such particles we may take the set of results for ²⁵²Cf spontaneous fission by Cosper et al. ⁽³⁴⁾ given in Table 11.

Tabl	е	11	
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Yields of charged particles emitted in ternary spontaneous fission of ²⁵²Cf

Particle	Yield per 10 ⁴ fissions
1 _H	$0.37 \pm 0.05$
2 _H	0.21 ± 0.01
³ H	2.14 $\pm 0.07$
³ He	$< 2.5 \times 10^{-3}$
⁴ He	33 <b>.</b> 4 ^{**}
⁶ He	$0.65 \pm 0.05$
8 _{He}	$0.02 \pm 0.002$
Li isotopes	0.04 ± 0.005
Be isotopes	$0.05 \pm 0.005$

*Value from ref. (35)

Cosper et al. measured their yields relative to that of ⁴He.

There is a considerable body of experimental data on ternary fission yields of all the particles in the literature, including some on those for masses above 20 whose existance is still disputed, but which, if present at all, only occur to the extent of a few per  $10^6$  fissions. However, from the point of view of the users of fission product nuclear data the only product

of real importance is  3 H. This is because the amount of tritium produced in an operating nuclear reactor may well be sufficient to become an environmental hazard. The present information about  3 H yields in neutron fission is summarised in Table 12.

## Table 12

3 H	yields	in	neutron	fission
	The second second second second second second second second second second second second second second second s	****	سترحا ومحافظها منجارها لتكبيرك ويتتباذ فيهومها	water and the second second second second second second second second second second second second second second

Fissioning nuclide	Neutron energy	³ H yield per 10 ⁴ fissions	Reference
233 _U	Thermal	$0.91 \pm 0.06^{a}$	43
		$0.86 \pm 0.07^{a}$	44
		1.11 ^a	45
235			
0	Thermal	0.5 - 1	36
		0.95 ± 0.08	37
		0.80 ± 0.10	38
		1.0	39
		0.85 ± 0.09	40
		1.09 ^b	41
		1.08 ^b	42
	Fast 200-800 KeV	2.2 → 1.8	40
²³⁹ Pu	Thermal	1.35 ± 0.13	35
		1.67 ± 0.08	46
		< 1.82	43
a Relative	to a value of 24.1	per 10 ⁴ fissions fo	4 (35) or He
b "	¹¹ ¹¹ ¹¹ 23.0	- 11 11 11 11 1	3 11

# 8.3 Gaps in the data and recommendations for further work

From the point of view of reactor users of the data the major lack is that there is only one series of measurements at fast reactor energies, and since these measurements (on  235 U) show that the  3 H yield is more than a factor of 2 higher at these energies than at thermal it would seem advisable to make similar measurements for other nuclides of importance to the reactor programme. Extreme accuracy is probably not needed, the present level of about  $\pm$  10% being quite adequate. The lack of any data for fissioning nuclides such as  232 Th and  238 U is probably not important enough to justify making measurements on these and other constituents of reactor fuels, since the variation of the yields from nuclide to nuclide is quite small.

From a narrow pragmatic standpoint it does not seem worth while conducting experiments on the other light particles to fill the many gaps in the data either, but we ought to realise that ternary fission has considerable importance in helping our understanding of the basic fission process and that fundamental experiments in this field should therefore be encouraged.

#### 9. CONCLUSIONS

The general conclusion which results from the survey give in this paper is that measurements of fission yields above thermal energies are at present inadequate to meet the needs of fast reactor designers and operators, and grossly inadequate as a source of information for a fundamental consideration of the fission process in detail. While more evaluation work is needed, the primary requirement is for more absolute measurements on many fissioning nuclides under clearly defined conditions regarding the neutrons used, the main effort being put on to those fission products of the fuel nuclides which are used for monitoring purposes; high accuracy is needed for these particular measurements.

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### APPENDIX 1

# TABLE 1

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					232	
Recommended	fast	fission	yields	for	Th (pe	er cent)

Mass No.	Crouch*	Lammer and Eder	Meek and Rider	Mean	Mass No.	Crouch	Lammer and Eder	Meek and Rider	Mean
77	0.011	0.010	0.013	0.011	116		(0, 25)	0.064	0.057
78	0.011	(0.035)	0.034	0.034	117	0.048	0.049	0.070	0.056
79		(0.08)	0.058	0.069	118	0.010	(0.05)	0.060	0.055
80		(0,2)	0.15	0.175	119		(0.05)	0.061	0.055
81		(0.4)	0.37	0.385	120		0.05	0.056	0.053
82		(1,0)	1.03	1.01	121	0.046	0.055	0.047	0.049
83	2.00	1.87	2.14	2.00	122		(0.04)	0.043	0.041
84	3.72	3.44	3.91	3.69	123	0.027	0.031	0.044	0.034
85	3.95	4.00	4.06	4.00	124		(0.03)	0.032	0.031
86	6.11	5.66	6.25	6.01	125	0.037	0.033	0.034	0.035
87	6.57	5.99	6.81	6.46	126		(0.05)	0.036	0.043
88	6.92	6.32	7.18	6.81	127	0.17	0.09	0.078	0.113
89	6.96	6.72	7.79	7.16	128		(0.18)	0.17	0.175
90	7.12	7.40	7.85	7.37	129		(0.36)	0.38	0.37
91	5.18	7.26	7.33	6.59	130		(0.8)	0.84	0.82
92		7.49	7.00	7.24	131	1.57	1.52	1.58	1.56
93	7.86	7.21	7.66	7.58	132	2.29	2.70	2.67	2.49
94		(6.23)	5.87	6.05	133	2.75	3.74	4.09	3.53
95	5.30	5.30	5.64	5.41	134	5.48	5.06	5.15	5.23
96	•	(4.8)	4.94	4.87	135	4.66	4.65	5.07	4.79
97	4.65	3.96	4.38	4.33	136	5.55	5.30	5.20	5.35
98		(3.4)	3.71	3.56	137	5.34	4.61	5.44	5.18
99	2.78	2.76	2.85	2.81	138		(6.02)	7.02	6.52
100	ł	(1.9)	1.07	1.45	139	7.30	7.38	6.53	7.13
101		(1.14)	0.64	0.89	140	7.65	8.31	7.81	7.86
102		(0.5)	0.37	0.43	141	7.44	7.28	7.32	7.36
103	0.15	0.146	0.16	0.15	142		(7.22)	7.98	7,60
104		(0.08)	0.086	0.083	143	6.80	7.12	6.89	6,90
105	0.072	0.05	0.04	0.054	144	7.59	7.66	7.50	7.56
106	0.043	0.041	0.053	0.046	145	5.52	5.78	5.37	5.56
107	1	(0.04)	0,030	0.035	146	4.73	4.95	4.60	4.76
108		(0.04)	0.055	0.047	147	2.96	2.97	3.05	2.99
109	0.050	0.041	0.052	0.048	148	2.08	2.18	2.02	2.09
110		(0.045)	0.063	0.054	149	1.22	1.44	1.23	1.30
111	0.054	0.045	0.078	0.059	150	1.04	1.09	0.99	1.04
112	0.057	0.062	0.088	0.069	151	0.46	0.41	0.40	0.42
113	0.045	0.06	0.073	0.059	152		(0.32)	0.082	?
114		(0.06)	0.070	0.065	153	0.22	0.21	0.19	0.21
115	0.050	0.067	0.063	0.057	154		0.06	0.017	0,038

*Crouch has given two tables of ²³²Th yields, one for yields measured in thermal reactors and one for yields measured in fast. Because it seems probable that any small real differences in the yields measured under these two conditions are masked by the large errors in the measurements, they have been averaged together.

Yields in brackets are interpolated by the evaluators.

# APPENDIX 1

# TABLE 2

# Recommended fast fission yields for 233U (per cent)

Mass No.	Crouch*
Mass No. 89 91 99 103 106 111 115 118 119 120 122 124 126 129 137 140 143	Crouch* 6.25 6.59 4.75 0.633 0.155 0.032 0.056 0.06 0.074 0.083 0.083 0.083 0.12 0.286 1.57 6.60 6.31 4.83
122	0.083
124	0.12
126	0.286
129	1.57
137	5.60
140	6.31
143	4.83
143	3.94
144	1.59
149	0.75
151	0.32
153	0.103
157	0.0115

* Crouch's is the only current evaluation for this muclide.

# APPENDIX 1

# TABLE 3

Recommended	fast	fission	yields	for	²³⁵ U(per	cent)

Mass No.	Crouch	Meek and Rider	Mean	Mass No.	Crouch	Meek and Rider	Mean
74 75 76 77 78 79 80 81 82 83 84 85 86 87 88 89 90 91 92 93 94 95 96 97 98 99 100 101 102 103 104 105 106 107 108 109 110 111 112 113 114 115 116	0.615 1.07 1.41 1.93 2.54 3.63 4.39 5.10 6.44 5.95 6.04 5.55 6.04 5.55 6.04 5.55 6.04 5.55 6.04 5.55 6.04 5.55 6.04 5.55 6.04 5.55 6.04 5.55 6.04 5.55 6.04 5.55 6.04 5.55 6.04 5.55 6.04 5.55 6.04 5.55 6.04 5.55 6.04 5.55 6.04 5.55 6.04 5.55 6.04 5.55 6.04 5.55 6.04 5.55 6.04 5.55 6.04 5.55 6.04 5.55 6.04 5.55 6.04 5.55 6.0437 0.028 0.034 0.034 0.022 0.036	R1der         0.0027         0.0093         0.029         0.045         0.077         0.086         0.18         0.29         0.42         0.63         1.43         1.86         2.56         3.49         4.81         5.25         5.40         5.69         5.96         6.06         6.27         6.28         5.91         5.82         6.11         5.82         6.11         5.82         6.11         5.10         4.49         3.10         2.27         1.40         0.97         0.35         0.24         0.12         0.11         0.033         0.033         0.033         0.033	0.622 1.05 1.42 1.89 2.55 3.56 4.60 5.17 6.35 5.93 5.93 5.93 5.68 6.23 5.28 4.57 3.20 2.31 1.42 0.703 0.035 0.035 0.033 0.033 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0.035 0	$\begin{array}{c} 117\\ 118\\ 19\\ 120\\ 121\\ 122\\ 123\\ 124\\ 125\\ 126\\ 127\\ 128\\ 129\\ 130\\ 131\\ 132\\ 133\\ 134\\ 135\\ 136\\ 137\\ 138\\ 139\\ 140\\ 141\\ 142\\ 143\\ 144\\ 145\\ 146\\ 147\\ 148\\ 149\\ 150\\ 151\\ 152\\ 153\\ 154\\ 155\\ 156\\ 157\\ 158\\ 159\\ \end{array}$	0.073 (1.4) 3.23 4.54 6.57 7.09 6.26 5.93 5.99 6.60 5.78 5.99 5.82 5.80 4.94 3.82 2.96 1.99 1.71 1.09 0.72 0.44 0.309 0.198 0.098 (0.035) 0.015	R10er         0.038         0.036         0.038         0.037         0.038         0.037         0.038         0.037         0.038         0.037         0.038         0.037         0.038         0.037         0.038         0.037         0.038         0.050         0.068         0.092         0.072         0.24         0.37         0.60         0.85         2.00         3.21         4.51         6.60         6.98         6.45         5.84         6.19         6.50         6.38         6.03         5.96         5.66         5.77         5.20         3.83         2.98         2.05         1.72         1.07         0.75         0.43         0.066         0.028         0.019         0	0.072 1.7 3.22 4.52 6.58 7.03 6.36 5.88 6.09 6.55 5.91 5.97 5.74 5.97 5.74 5.97 5.74 5.97 5.74 5.07 3.82 2.97 2.02 1.71 1.08 0.735 0.435 0.304 0.194 0.097 0.050 0.022 0.0031
				טסו		0.0014	
### TABLE 4

# Recommended fast fission yields for ²³⁸U (per cent)

Mass No.	Crouch	Meek and Rider	Mean	Mass No.	Crouch	Meek and Rider	Mean
Mass No. 77 78 79 80 81 82 83 84 85 86 87 88 89 90 91 92 93 94 95 96 97 98 99 100 101 102 103 104 105 106 107 108 109 110 111	Crouch 0.0036 3.16 3.14 5.54 5.54 5.89 6.27 6.01 3.50 2.90 0.13 0.0774	Meek and Rider 0.0038 0.0143 0.0411 0.088 0.16 0.26 0.41 0.85 0.81 1.36 1.42 1.68 3.02 3.29 4.51 3.91 4.83 5.14 5.58 5.49 5.96 6.03 6.42 6.37 6.38 6.38 6.40 4.51 3.23 2.84 1.31 0.64 0.27 0.14 0.10	Mean 0.0037 3.09 3.21 5.56 5.92 6.35 6.20 3.37 2.87 0.20 0.0887	$\begin{array}{c} \text{Mass No.}\\ 119\\ 120\\ 121\\ 122\\ 123\\ 124\\ 125\\ 126\\ 127\\ 128\\ 129\\ 130\\ 131\\ 132\\ 133\\ 134\\ 135\\ 136\\ 137\\ 138\\ 139\\ 140\\ 141\\ 142\\ 143\\ 144\\ 145\\ 146\\ 147\\ 148\\ 149\\ 150\\ 151\\ 152\\ 153\\ \end{array}$	Crouch 0.078 0.154 0.26 3.62 4.67 7.13 6.08 5.21 4.60 4.25 3.94 2.64 2.40 1.78 1.49 0.43	Meek and Rider 0.037 0.038 0.039 0.042 0.046 0.113 0.064 0.107 0.31 0.65 1.47 3.66 5.33 6.47 7.53 6.67 6.76 5.95 5.92 5.27 5.95 5.45 4.70 4.53 4.54 3.74 3.87 2.57 2.12 1.84 1.29 0.93 0.58 0.41	Mean 0.096 0.131 0.455 3.64 5.00 6.54 6.02 4.87 4.57 4.00 3.90 2.60 2.26 1.81 1.39 0.42
110 111 112 113	0.0774 0.07	0.14 0.10 0.089 0.055	0.0887 0.079	152 153 154 155	0.43	0.58 0.41 0.24 0.14	0.42
114 115 116 117	0.0407	0.042 0.053 0.038 0.039	0.0468	156 157 158 159	0,067 0,0083	0.075 0.038 0.017 0.0088	0.071
118		0.037		160 161	0.0015	0.0034 0.0020	0.0017

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### TABLE 5

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Recommended fast fission yields for <sup>237</sup>Np (per cent)
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Mass No.	Von Gunten	Crouch	Mean
Mass No. 83 89 91 93 95 97 99 103 105 106 109 111 112 113 115 121 125 127 129 131 132 133 135 140 141 143 144 147 149	Von Gunten 0.43 2.00 4.60 5.72 5.70 5.87 6.92 5.48 3.23 2.06 0.45 0.103 0.063 0.057 0.048 0.078 0.117 0.47 3.18 3.47 5.82 6.42 5.35 5.93 5.49 5.54 3.89 3.21 1.88 1.07	Crouch 5.97 5.96 6.05 6.90 0.110 3.20 6.12 6.29 5.52 6.36 5.70 4.54 2.64 1.74	Mean 0.43 2.00 4.60 5.84 5.83 5.96 6.91 5.48 3.23 2.06 0.45 0.106 0.063 0.057 0.048 0.078 0.117 0.47 3.18 3.33 5.97 6.35 5.43 5.93 6.42 5.62 4.21 2.92 1.81
153 156	0.44 0.23	0.442	0.44 0.23

## TABLE 6

Mass No.	Crouch	Meek and Rider	Mean	Mass No.	Crouch	Meek and Rider	Mean
77 78 79 80	0.0134 0.035	0.015 0.038 0.068	0.0142 0.0365	124 125 126	0.19	0.088 0.192 0.304 0.302	0,191
81 82 83 84	0.36 0.56	0.172 0.250 0.350 0.547	0.355	128 129 130 131	1.17 4.45	0.80 0.92 2.08 4.20	1.04 4.32
85 86 87 88	0.657 0.882 1.16 1.44	0.642 0.84 1.11 1.38	0.657 0.861 1.135 1.41	132 133 134 135	5.42 6.91 7.35 7.54	5.37 6.82 7.26 7.45	5.39 6.86 7.30 7.49
89 90 91 92	1.87 2.18 2.58 3.13	1.72 2.09 2.46 2.99	1.79 2.13 2.52 3.06	136 137 138 139	6.92 6.69 4.97 6.11	6.83 6.63 4.92 6.84	6.87 6.66 4.94 6.47
93 94 95 96 97	3.91 4.40 4.84 5.11	3.73 4.19 4.59 4.88	3.82 4.29 4.71 4.99	140 141 142 143	$5 \cdot 26$ $5 \cdot 84$ $4 \cdot 95$ $4 \cdot 45$ $7 \cdot 51$	5.50 6.09 4.89 4.31 7.61	5.38 5.96 4.92 4.38 3.56
98 99 100 101	5.81 5.81 6.76 6.88	5.21 5.55 5.61 6.46 6.58	5.68 5.71 6.61 6.73	145 146 147 148	3.05 2.52 1.73	3.02 2.50 2.03 1.69	3.035 2.51 1.71
102 103 104 105	6.97 6.79 6.77	6.58 6.53 6.48 5.12	6.77 6.66 6.62	149 150 151 152	1.36 1.05 0.84 0.683	1.33 1.03 0.82 0.67	1.34 1.04 0.83 0.676
106 107 108 109	4.82 1.90	4.52 3.58 2.61 1.62	4.67 1.76	153 154 155 156	0.51 0.324 0.159	0.481 0.317 0.238 0.157	0.495 0.320 0.158
111 112 113 114	0.439 0.137 0.0915 0.099	0.79 0.37 0.20 0.127 0.094	0.404 0.168 0.109 0.096	158 159 160 161	0.108	0.071 0.044 0.025 0.014	0.111
115 116 117 118	0.115 0.064	0,089 0.061 0.084 0.083	0.102 0.062				
119 120 121 122 123	0.0169	0.066 0.064 0.066 0.071 0.084	0.0414				

# Recommended fast fission yields for 239Pu (per cent)

#### TABLE 7

Comparison of recommended fast fission yields for

²³²Th as evaluated by Lammer in Ref. 2 (included

#### in Appendix 1 Table 1) and in Ref. 70

Mass No.	Ref. 70	Ref. 2	Mass No.	Ref. 70	Ref. 2
Mass No. 72 73 77 83 84 85 86 87 88 89 90 91 93 95 97 97 99 103 105 106 109	Ref. 70 0.00034 0.00046 0.01 1.90 3.44 3.74 5.69 6.01 6.34 6.43 7.27 6.92 7.65 5.41 4.02 2.74 0.153 0.04 0.041 0.040	Ref. 2 0.0102 1.87 3.44 4.02 5.66 5.99 6.32 6.72 7.40 7.26 7.21 5.30 3.96 2.76 0.146 0.05 0.041 0.042	Mass No. 121 123 125 127 131 132 133 134 135 136 137 139 140 141 143 144 145 146 147 148	Ref. 70 0.056 0.032 0.034 0.076 1.52 2.69 3.75 5.06 4.76 5.38 4.50 6.78 8.45 7.50 7.02 7.49 5.70 4.88 3.13 2.15	Ref. 2 0.055 0.031 0.033 0.089 1.52 2.70 3.74 5.06 4.65 5.30 4.44 7.38 8.31 7.28 7.12 7.66 5.78 4.95 2.97 2.18
111 112 113 115 117	0,04 0,06 0,058 0,057 0,051	0.045 0.062 0.06 0.057 0.049	149 150 151 153 156	1.47 1.08 0.42 0.21 0.0026	1.44 1.09 0.41 0.21 0.0026

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### TABLE 1

Mass No.	Von Gunten	Crouch	Mean
66		0.000131	0.000131
67	ĺ	0.00026	0.00026
72		0.007	0.007
73		0.0076	0.0076
77	1	0,124	0.124
78		0,295	0, 295
79		0.903	0,903
81	1	1.15	1.15
83	1.52	1.60	1.56
84	1.86	2.08	1.97
88		4.28	4.28
89	5.7	5.81	5.75
90		5.54	5.54
91	5.88	5.50	5.69
92		5.41	5.41
93	5.78	5.60	5.69
95	6.7	6.70	6,70
97	3.8	3.32	3.56
99	1.92	1.90	1.91
101	1.60	1.52	1.56
102	0.70	0.67	0.68
103	0.75	0.794	0.77
105	1.06	1.02	1.04
106	1.07	1.07	1.07
109	1.10	1.14	1.72
110	1.42	1.22	1.32
112	1.29	1.29	1.29
115	3.20	1.18	1.20
115	1.20	0.01	1.35
121	1.0	0.915	0.50
123	0.00	0.30	1.01
120	1.10		1.21
129	1.19	2.05	2.05
170	0.69	2.03	2.03
132	2:00	2,10	2.73
174		3.13 6 46	5.19
175		0.49	0.49
170	6.02	4.00 5 61	5 68
140	0.02	5 80	5.00
140	FOF	5.00	5.00
141	0.00 7.77	3.50	0+0F # 70
140	0.00 5.10	0.20	0.00
145	5.12	2:01	5.06
140	5.00		3.00

Recommend 14 MeV fission yields for 232 Th (per cent)

TABLE 1	(cont'	d)
	-	

Mass Nc.	Von Gunten	Crouch	Mean
147 149 151 153 156 157 159 161 166 169	1.70 0.66 0.16 0.085 0.036 0.012	1.81 0.086 0.0044 0.0016 0.000029 0.000023	1.75 0.66 0.16 0.086 0.036 0.012 0.0044 0.0016 0.000029 0.000023

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### TABLE 2

Recommended 14 MeV fission yields for 233 (per cent)

Mass No.	Crouch*
66	0.00077
67	0.0018
72	0.0146
83	1.33
84	2.02
89	4.82
91	5.40
92	5.72
93	6,00
95	5.60
97	5.20
99	3.69
103	2.31
105	1.88
106	1.52
109	1.20
111	1.27
112	1.46
113	1.06
115	1.31
121	1.06
125	1.51
127	2.10
131	3.4
132	3.82
133	4.37
134	4.05
177	4.90
137	5 70
140	0 - 1 0 A 27
14.1	4.70
143	3.60
144	2.60
147	1.29
153	0.156
159	0.0116
161	0.005

*Crouch's is the only current evaluation of this nuclide.

### TABLE 3

Recommended 14 MeV fission yields for  $235_{U}$  (per cent)

Mass No.	von Gunten	Meek & Rider	Crouch	Mean
66		0.00030	0.00034	0.00032
67		0.00064	0.00065	0.00065
68		0.00088	(0,001)	0,00094
69		0.00138	(0.0016)	0.00149
70		0.00236	(0.0026)	0.00248
71		0.00393	(0.004)	0.00397
72		0.00586	0.0067	0.00628
73		0.0107	(0.0085)	0.0092
74		0.0167	(0.017)	0.0169
75		0.0265	(0.027)	0.0267
76		0.0393	(0.043)	0.0411
77		0.0631	0.069	0.066
78		0.0983	(0.105)	0.102
79		0.163	(0.17)	0.166
80		0.190	(0.255)	0.222
81		0.250	0.362	0.306
82		0.379	(0.62)	0.499
83	1.23	1.01	0.97	1.07
84		1.21	1.05	1.13
85		2.00	(1.5)	1.75
86		2.46	(2.0)	2.23
87		2.96	(2.7)	2.83
88	}	3.52	(3.5)	3.51
89	4.16	3.95	4.31	4.14
90	4.5	4.46	4.4	4.45
91	4.71	4.47	4.98	4.72
92		4.88		4.88
93	1	5.30	5.40	5.35
94		4.80		4.80
95	4.80	4.85	4.71	4.79
96		4.85		4.85
97	5.29	5.18	5.24	5.24
98	F 0F	4.10	= 0	4.70
99	5.25	0.10	3.20	3,23
100		4.00	1	7.01
100		3.34		3.54
102	2 25	3 42	3 40	3 70
104	0.00	2.30	0,10	2.30
105	2.00	2 07	2 12	2.08
100	2.09	1 64	1 75	1 70
107	1.70	1 79	1040	1.78
107		1 37	1	1.33
100		1,00		
L	1			L

$ \begin{array}{ c c c c c c c c c c c c c c c c c c c$	Mass No.	von Gunten	Meek & Rider	Crouch	Mean
$ \begin{array}{ c c c c c c c c c c c c c c c c c c c$	109	1.17	1.17	1.47	1.27
$ \begin{array}{ c c c c c c c c c c c c c c c c c c c$	110		1.18		1.18
$ \begin{array}{ c c c c c c c c c c c c c c c c c c c$	111	1.11	1.14	1.10	1.12
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	112	0.93	0,907	0.89	0.91
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	113	1.15	1.25	0.922	1.11
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	114		1.03		1.03
1161.021.021171.301.301181.061191.071201.111211.091.231.231221.231231.231241.321251.451261.712.092.074.332.831282.751292.42.42.871314.234.004.194.595.145.75.615.65.615.75.614.754.755.95.415.95.415.95.415.95.255.91364.754.751404.614.624.474.513.813.813.853.813.813.813.813.813.813.813.813.813.813.813.813.853.873.843.813.853.873.813.813.853.873.843.853.873.843.853.873.843.853.873.813.853.873.883.813.85 <t< td=""><td>115</td><td>0.97</td><td>0.943</td><td>1.12</td><td>1.01</td></t<>	115	0.97	0.943	1.12	1.01
$ \begin{array}{c ccccccccccccccccccccccccccccccccccc$	116		1.02		1.02
1181.061.061191.071.071201.111.071201.111.111211.091.081221.231.231231.231.231241.321.321251.451.281261.713.202.452.751292.42.872.63303.623114.234.704.714.374.595.65.615.75.614.754.751335.65.75.614.754.751385.251395.04.964.834.064.33.813.813.853.813.853.813.853.813.854.624.474.614.624.474.571443.203.523.693.613.623.613.623.623.614.624.754.754.751404.614.624.474.571433.813.853.623.523.693.523.693.523.693.533.613.54<	117		1.30		1.30
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	118		1.06		1.06
120 $1.11$ $1.11$ $1.11$ $1.11$ $121$ $1.09$ $1.08$ $1.025$ $1.06$ $122$ $1.23$ $1.23$ $1.23$ $123$ $1.23$ $1.23$ $1.23$ $124$ $1.32$ $1.32$ $1.32$ $125$ $1.45$ $1.28$ $1.91$ $1.55$ $126$ $1.71$ $3.20$ $2.45$ $127$ $2.09$ $2.07$ $4.33$ $2.83$ $128$ $2.75$ $2.75$ $2.75$ $129$ $2.4$ $2.87$ $2.63$ $130$ $3.62$ $3.62$ $131$ $4.23$ $4.00$ $4.19$ $4.14$ $3.29$ $3.62$ $133$ $5.6$ $5.61$ $5.56$ $5.9$ $5.18$ $5.14$ $5.41$ $135$ $5.7$ $5.61$ $4.55$ $5.29$ $136$ $4.75$ $4.75$ $4.75$ $138$ $5.9$ $5.41$ $5.90$ $5.74$ $138$ $5.0$ $4.96$ $4.83$ $4.93$ $140$ $4.61$ $4.62$ $4.47$ $4.57$ $141$ $3.8$ $4.52$ $3.80$ $4.04$ $142$ $2.94$ $2.94$ $2.94$ $146$ $2.47$ $2.47$ $2.47$ $145$ $0.548$ $0.548$ $0.548$ $150$ $0.548$ $0.548$ $0.548$ $150$ $0.724$ $0.204$ $0.204$ $155$ $0.08$ $0.0566$ $0.062$ $0.079$	119		1.07		1.07
121 $1.09$ $1.08$ $1.025$ $1.06$ 122 $1.23$ $1.23$ $1.23$ $1.23$ 123 $1.23$ $1.23$ $1.23$ 124 $1.32$ $1.32$ 125 $1.45$ $1.28$ $1.91$ 126 $1.71$ $3.20$ $2.45$ 127 $2.09$ $2.07$ $4.33$ $2.83$ 128 $2.75$ $2.63$ $3.62$ 130 $3.62$ $3.62$ $3.62$ 131 $4.23$ $4.00$ $4_019$ $4.14$ $3.20$ $4.71$ $4_037$ $4.59$ $5.18$ $5.14$ $5.41$ $135$ $5.7$ $5.61$ $4.55$ $5.29$ $134$ $5.9$ $5.41$ $5.90$ $5.74$ $138$ $5.25$ $5.25$ $5.25$ $139$ $5.0$ $4.96$ $4.83$ $140$ $4.61$ $4.62$ $4.47$ $4.51$ $3.81$ $3.85$ $3.87$ $141$ $3.81$ $3.85$ $3.87$ $144$ $3.20$ $3.52$ $3.09$ $2.94$ $2.47$ $2.47$ $144$ $0.514$ $0.548$ $150$ $0.314$ $0.314$ $0.548$ $0.966$ $0.962$ $0.966$ $0.962$ $0.979$	120		1.11		1.11
122 $1.23$ $1.23$ $1.23$ $125$ $1.45$ $1.23$ $1.32$ $125$ $1.45$ $1.28$ $1.91$ $125$ $1.45$ $1.28$ $1.91$ $127$ $2.09$ $2.07$ $4.33$ $128$ $2.75$ $2.83$ $129$ $2.4$ $2.87$ $2.63$ $130$ $3.62$ $3.62$ $131$ $4.23$ $4.00$ $4.19$ $132$ $4.70$ $4.71$ $4.37$ $4.37$ $4.59$ $133$ $5.6$ $5.61$ $5.7$ $5.61$ $4.55$ $5.9$ $5.18$ $5.14$ $5.9$ $5.41$ $5.90$ $5.7$ $5.61$ $4.55$ $5.91$ $5.9$ $5.41$ $5.92$ $5.74$ $5.9$ $5.41$ $5.90$ $5.74$ $5.25$ $5.93$ $5.0$ $4.96$ $4.83$ $4.93$ $140$ $4.61$ $4.62$ $4.47$ $4.57$ $141$ $3.8$ $3.85$ $3.81$ $3.85$ $3.87$ $3.84$ $4.06$ $143$ $3.81$ $3.85$ $3.94$ $4.06$ $144$ $0.204$ $0.204$ $0.548$ $0.314$ $0.314$ $0.548$ $0.314$ $0.314$ $151$ $0.096$ $(0.44)$ $0.153$ $0.224$ $0.242$ $154$ $0.096$ $(0.085)$ $0.096$ $0.062$ $0.079$	121	1.09	1.08	1.025	1,06
$ \begin{array}{c ccccccccccccccccccccccccccccccccccc$	122		1.23		1.23
124 $1.32$ $1.32$ $1.32$ $125$ $1.45$ $1.28$ $1.91$ $1.55$ $126$ $1.71$ $3.20$ $2.45$ $127$ $2.09$ $2.07$ $4.33$ $2.83$ $128$ $2.75$ $2.75$ $2.75$ $129$ $2.4$ $2.87$ $2.63$ $130$ $3.62$ $3.62$ $3.62$ $131$ $4.23$ $4.00$ $4.19$ $4.14$ $132$ $4.70$ $4.71$ $4.377$ $4.59$ $133$ $5.6$ $5.61$ $5.56$ $5.99$ $134$ $5.9$ $5.18$ $5.14$ $5.41$ $135$ $5.7$ $5.61$ $4.55$ $5.29$ $136$ $4.75$ $4.75$ $5.25$ $139$ $5.0$ $4.96$ $4.83$ $4.93$ $140$ $4.61$ $4.62$ $4.477$ $4.57$ $141$ $3.81$ $3.85$ $3.87$ $3.84$ $144$ $3.20$ $3.52$ $3.09$ $3.27$ $145$ $2.94$ $2.94$ $2.94$ $146$ $2.47$ $2.47$ $2.47$ $148$ $1.37$ $1.37$ $1.37$ $149$ $0.548$ $0.548$ $0.548$ $150$ $0.314$ $0.204$ $0.204$ $0.204$ $0.204$ $0.204$ $0.204$ $152$ $0.153$ $0.724$ $(0.085)$ $0.096$ $0.096$ $(0.14)$ $0.112$ $155$ $0.08$ $0.0966$ $0.0258$	123		1.23		1.23
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	124		1.32		1.32
1261.71 $3.20$ $2.45$ 127 $2.09$ $2.07$ $4.33$ $2.83$ 128 $2.75$ $2.75$ $2.75$ 129 $2.4$ $2.87$ $2.63$ 130 $3.62$ $3.62$ 131 $4.23$ $4.00$ $4.19$ $4.71$ $4.37$ $4.59$ 133 $5.6$ $5.61$ $5.7$ $5.61$ $5.56$ $5.9$ $5.18$ $5.14$ $5.9$ $5.18$ $5.14$ $5.7$ $5.61$ $4.55$ $5.9$ $5.41$ $5.90$ $5.74$ $5.90$ $5.74$ $138$ $5.25$ $5.25$ $139$ $5.0$ $4.96$ $4.83$ $4.93$ $140$ $4.61$ $4.62$ $4.47$ $4.57$ $141$ $3.8$ $3.85$ $3.80$ $4.04$ $142$ $4.06$ $143$ $3.81$ $3.81$ $3.85$ $3.87$ $3.84$ $144$ $2.94$ $2.94$ $2.94$ $146$ $2.47$ $1.37$ $0.548$ $0.548$ $0.548$ $0.548$ $0.548$ $0.548$ $0.204$ $0.204$ $0.204$ $152$ $0.724$ $0.096$ $(0.112)$ $0.5566$ $0.062$ $0.068$ $0.0666$	125	1.45	1.28	1.91	1.55
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	126		1.71	3.20	2.45
1282.752.751292.42.872.63130 $3.62$ $3.62$ 131 $4.23$ $4.00$ $4.19$ 132 $4.70$ $4.71$ $4.37$ $4.59$ $5.6$ $5.61$ $5.56$ 133 $5.6$ $5.61$ $5.56$ 134 $5.9$ $5.18$ $5.14$ 135 $5.7$ $5.61$ $4.55$ 136 $4.75$ $4.75$ 137 $5.9$ $5.41$ $5.90$ 138 $5.25$ $5.25$ 139 $5.0$ $4.96$ $4.83$ 140 $4.61$ $4.62$ $4.47$ 141 $3.8$ $4.52$ $3.80$ 144 $3.20$ $3.52$ $3.09$ 145 $2.94$ $2.94$ 146 $2.47$ $2.47$ 147 $1.70$ $1.65$ 153 $0.204$ $0.204$ $0.548$ $0.548$ $0.548$ $150$ $0.153$ $0.153$ $0.204$ $0.204$ $0.204$ $152$ $0.066$ $(0.14)$ $0.112$ $0.5566$ $0.066$ $0.062$ $0.058$	127	2.09	2.07	4.33	2.83
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	128		2.75		2.75
130 $3.62$ $3.62$ 131 $4.23$ $4.00$ $4.19$ $4.14$ 132 $4.70$ $4.71$ $4.37$ $4.59$ 133 $5.6$ $5.61$ $5.56$ $5.59$ 134 $5.9$ $5.18$ $5.14$ $5.41$ 135 $5.7$ $5.61$ $4.55$ $5.29$ 136 $4.75$ $4.75$ $4.75$ 137 $5.9$ $5.41$ $5.90$ $5.74$ 138 $5.25$ $5.25$ $5.25$ 139 $5.0$ $4.96$ $4.83$ $4.93$ 140 $4.61$ $4.62$ $4.47$ $4.57$ 141 $3.8$ $3.85$ $3.87$ $3.84$ 142 $4.06$ $4.06$ $4.06$ 143 $3.81$ $3.85$ $3.87$ $3.84$ 144 $3.20$ $3.52$ $3.09$ $3.27$ 145 $2.94$ $2.94$ $2.94$ 146 $2.47$ $2.47$ $1.65$ 150 $0.314$ $0.314$ $0.314$ 151 $0.204$ $0.204$ $0.204$ 152 $0.153$ $0.724$ $0.242$ 154 $0.096$ $(0.14)$ $0.112$ 155 $0.724$ $(0.085)$ $0.079$ 156 $0.08$ $0.2666$ $0_0062$ $0.058$	129	2.4	2.87		2.63
131 $4.23$ $4.00$ $4_{0}19$ $4.14$ 132 $4.70$ $4.71$ $4_{0}37$ $4.59$ 133 $5.6$ $5.61$ $5.56$ $5.59$ 134 $5.9$ $5.18$ $5.14$ $5.41$ 135 $5.7$ $5.61$ $4.55$ $5.29$ 136 $4.75$ $4.75$ $4.75$ 137 $5.9$ $5.41$ $5.90$ 138 $5.25$ $5.25$ 139 $5.0$ $4.96$ $4.83$ 140 $4.61$ $4.62$ $4.47$ 141 $3.8$ $4.52$ $3.80$ 142 $4.06$ $4.06$ 143 $3.81$ $3.85$ $3.87$ 3.81 $3.85$ $3.87$ $3.84$ 144 $3.20$ $3.52$ $3.09$ $3.27$ 145 $2.94$ $2.94$ $2.94$ 146 $2.477$ $1.65$ $1.67$ 148 $0.314$ $0.204$ $0.204$ 151 $0.204$ $0.204$ $0.204$ 152 $0.153$ $0.242$ $0.242$ 154 $0.096$ $(0.14)$ $0.112$ 155 $0.08$ $0.0566$ $0.062$ $0.056$ $0.062$ $0.058$	130		3.62		3.62
1324.704.71 $4.37$ $4.59$ 1335.65.615.565.591345.95.185.145.411355.75.614.555.291364.754.754.751375.95.415.905.741385.255.255.251395.04.964.834.931404.614.624.474.571413.84.523.804.041424.064.064.061433.813.853.873.813.853.873.841443.203.523.093.271452.942.472.471471.371.651.671480.5480.3140.5481500.5480.3140.2041510.2040.2041520.1530.2421540.096(0.14)1550.080.05660.0620.058	131	4.23	4.00	4.19	4.14
1335.65.615.565.591345.95.185.145.411355.75.61 $4.55$ 5.29136 $4.75$ $4.75$ $4.75$ 1375.95.415.90138 $5.25$ $5.25$ 1395.0 $4.96$ $4.83$ 140 $4.61$ $4.62$ $4.47$ 141 $3.8$ $4.52$ $3.80$ 142 $4.06$ $4.06$ 143 $3.81$ $3.85$ $3.87$ 144 $3.20$ $3.52$ $3.09$ 145 $2.94$ $2.94$ 146 $2.47$ $1.65$ 150 $0.548$ $0.548$ 150 $0.314$ $0.314$ 151 $0.204$ $0.204$ 152 $0.153$ $0.153$ 153 $0.242$ $0.242$ 154 $0.096$ $(0.14)$ $0.566$ $0.062$ $0.058$	132	4.70	4.71	4.37	4.59
134 $5.9$ $5.18$ $5.14$ $5.41$ 135 $5.7$ $5.61$ $4.55$ $5.29$ 136 $4.75$ $4.75$ 137 $5.9$ $5.41$ $5.90$ 138 $5.25$ $5.25$ 139 $5.0$ $4.96$ $4.83$ 140 $4.61$ $4.62$ $4.47$ 141 $3.8$ $4.52$ $3.80$ 142 $4.06$ $4.06$ 143 $3.81$ $3.85$ $3.87$ 3.81 $3.85$ $3.87$ $3.84$ 144 $3.20$ $3.52$ $3.09$ 145 $2.94$ $2.94$ 146 $2.477$ $2.477$ 147 $1.70$ $1.65$ $1.67$ 148 $1.37$ $0.548$ $0.548$ 150 $0.314$ $0.314$ $0.314$ 151 $0.204$ $0.204$ $0.204$ 152 $0.153$ $0.153$ $0.153$ 153 $0.242$ $0.242$ $0.242$ 154 $0.096$ $(0.14)$ $0.112$ 156 $0.08$ $0.0566$ $0.062$ $0.058$	133	5.6	5.61	5.56	5.59
135 $5.7$ $5.61$ $4.55$ $5.29$ 136 $4.75$ $4.75$ $4.75$ 137 $5.9$ $5.41$ $5.90$ $5.74$ 138 $5.25$ $5.25$ $5.25$ 139 $5.0$ $4.96$ $4.83$ $4.93$ 140 $4.61$ $4.62$ $4.47$ $4.57$ 141 $3.8$ $4.52$ $3.80$ $4.04$ 142 $4.06$ $4.06$ $4.06$ 143 $3.81$ $3.85$ $3.87$ $3.84$ 144 $3.20$ $3.52$ $3.09$ $3.27$ 145 $2.94$ $2.94$ $2.94$ 146 $2.477$ $2.47$ 147 $1.70$ $1.65$ $1.67$ 148 $0.548$ $0.548$ $0.548$ 150 $0.314$ $0.314$ $0.314$ 151 $0.204$ $0.204$ $0.204$ 152 $0.153$ $0.153$ $0.153$ 153 $0.242$ $0.242$ $0.242$ 154 $0.096$ $(0.14)$ $0.112$ 155 $0.08$ $0.0566$ $0_0062$ $0.058$	134	5.9	5.18	5.14	5.41
136 $4,75$ $4.75$ 137 $5.9$ $5.41$ $5.90$ $5.74$ 138 $5.25$ $5.25$ $5.25$ 139 $5.0$ $4.96$ $4.83$ $4.93$ 140 $4.61$ $4.62$ $4.47$ $4.57$ 141 $3.8$ $4.52$ $3.80$ $4.04$ 142 $4.06$ $4.06$ $4.06$ 143 $3.81$ $3.85$ $3.87$ $3.84$ 144 $3.20$ $3.52$ $3.09$ $3.27$ 145 $2.94$ $2.94$ $2.94$ 146 $2.477$ $1.65$ $1.67$ 148 $1.37$ $1.37$ $1.37$ 149 $0.548$ $0.548$ $0.548$ 150 $0.314$ $0.314$ $0.314$ 151 $0.204$ $0.204$ $0.204$ 152 $0.153$ $0.153$ $0.153$ 153 $0.242$ $0.242$ $0.242$ 154 $0.096$ $(0.14)$ $0.112$ 155 $0.08$ $0.0566$ $0_0062$ $0.058$	135	5.7	5.61	4.55	5.29
137 $5.9$ $5.41$ $5.90$ $5.74$ 138 $5.25$ $5.25$ $5.25$ $5.25$ 139 $5.0$ $4.96$ $4.83$ $4.93$ 140 $4.61$ $4.62$ $4.47$ $4.57$ 141 $3.8$ $4.52$ $3.80$ $4.04$ 142 $4.06$ $4.06$ $4.06$ 143 $3.81$ $3.85$ $3.87$ $3.80$ $4.06$ $4.06$ 144 $3.20$ $3.52$ $3.09$ $3.27$ $2.94$ $2.94$ $146$ $2.477$ $2.477$ $147$ $1.70$ $1.65$ $1.67$ $148$ $0.314$ $0.314$ $150$ $0.314$ $0.314$ $0.204$ $0.204$ $0.204$ $152$ $0.096$ $(0.14)$ $0.112$ $154$ $0.096$ $(0.14)$ $0.112$ $155$ $0.08$ $0.0566$ $0.062$ $0.058$	136		4.75		4.75
138 $5.25$ $5.25$ 139 $5.0$ $4.96$ $4.83$ $4.93$ 140 $4.61$ $4.62$ $4.47$ $4.57$ 141 $3.8$ $4.52$ $3.80$ $4.04$ 142 $4.06$ $4.06$ $4.06$ 143 $3.81$ $3.85$ $3.87$ 144 $3.20$ $3.52$ $3.09$ 145 $2.94$ $2.94$ 146 $2.47$ $2.47$ 147 $1.70$ $1.65$ 148 $0.548$ $0.548$ 150 $0.314$ $0.314$ 0.548 $0.548$ $0.548$ 151 $0.204$ $0.204$ 152 $0.153$ $0.153$ 153 $0.242$ $0.242$ 154 $0.096$ $(0.14)$ $0.566$ $0_0062$ $0.058$	137	5.9	5.41	5.90	5.74
139 $5.0$ $4.96$ $4.83$ $4.93$ 140 $4.61$ $4.62$ $4.47$ $4.57$ 141 $3.8$ $4.52$ $3.80$ $4.04$ 142 $4.06$ $4.06$ $4.06$ 143 $3.81$ $3.85$ $3.87$ $3.84$ 144 $3.20$ $3.52$ $3.09$ $3.27$ 145 $2.94$ $2.94$ $2.94$ 146 $2.47$ $1.65$ $1.67$ 148 $1.37$ $1.37$ $1.37$ 149 $0.548$ $0.548$ $0.548$ 150 $0.314$ $0.204$ $0.204$ 152 $0.153$ $0.153$ $0.153$ 153 $0.242$ $0.242$ $0.242$ 154 $0.096$ $(0.14)$ $0.112$ 155 $0.08$ $0.0566$ $0.062$ $0.058$	138		5.25		5.25
140 $4.61$ $4.62$ $4.47$ $4.57$ 141 $3.8$ $4.52$ $3.80$ $4.04$ 142 $4.06$ $4.06$ $4.06$ 143 $3.81$ $3.85$ $3.87$ 144 $3.20$ $3.52$ $3.09$ 145 $2.94$ $2.94$ 146 $2.47$ $2.47$ 147 $1.70$ $1.65$ 148 $1.37$ $1.37$ 149 $0.548$ $0.548$ 150 $0.314$ $0.314$ 0.153 $0.204$ $0.204$ 152 $0.153$ $0.153$ 153 $0.242$ $0.242$ 154 $0.096$ $(0.14)$ 0.566 $0.062$ $0.058$	139	5.0	4.96	4.83	4.93
141 $3.8$ $4.52$ $3.80$ $4.04$ 142 $4.06$ $4.06$ 143 $3.81$ $3.85$ $3.87$ $3.84$ 144 $3.20$ $3.52$ $3.09$ $3.27$ 145 $2.94$ $2.94$ $2.94$ 146 $2.47$ $2.47$ 147 $1.70$ $1.65$ $1.67$ 148 $1.37$ $0.548$ $0.548$ 150 $0.314$ $0.314$ $0.314$ 151 $0.204$ $0.204$ $0.204$ 152 $0.153$ $0.153$ $0.153$ 153 $0.242$ $0.242$ $0.242$ 154 $0.096$ $(0.14)$ $0.112$ 155 $0.08$ $0.0566$ $0.062$ $0.058$	140	4.61	4.62	4.47	4.57
142 $4.06$ $4.06$ $143$ $3.81$ $3.85$ $3.87$ $3.84$ $144$ $3.20$ $3.52$ $3.09$ $3.27$ $145$ $2.94$ $2.94$ $2.94$ $146$ $2.47$ $2.47$ $147$ $1.70$ $1.65$ $1.67$ $148$ $1.37$ $1.37$ $149$ $0.548$ $0.548$ $150$ $0.314$ $0.314$ $151$ $0.204$ $0.204$ $152$ $0.153$ $0.153$ $153$ $0.242$ $0.242$ $154$ $0.096$ $(0.14)$ $0.155$ $0.079$ $156$ $0.08$ $0.0566$ $0.062$ $0.058$	341	3.8	4.52	3.80	4.04
143 $3.81$ $3.85$ $3.87$ $3.84$ 144 $3.20$ $3.52$ $3.09$ $3.27$ 145 $2.94$ $2.94$ $2.94$ 146 $2.47$ $2.47$ 147 $1.70$ $1.65$ $1.67$ 148 $1.37$ $0.548$ $0.548$ 150 $0.314$ $0.314$ $0.314$ 151 $0.204$ $0.204$ $0.204$ 152 $0.153$ $0.242$ $0.242$ 154 $0.096$ $(0.14)$ $0.112$ 155 $0.08$ $0.0566$ $0.062$ $0.058$	142		4.06		4.06
144 $3.20$ $3.52$ $3.09$ $3.27$ $145$ $2.94$ $2.94$ $2.94$ $146$ $2.47$ $2.47$ $147$ $1.70$ $1.65$ $148$ $1.37$ $1.37$ $149$ $0.548$ $0.548$ $150$ $0.314$ $0.314$ $151$ $0.204$ $0.204$ $152$ $0.153$ $0.153$ $153$ $0.242$ $0.242$ $154$ $0.096$ $(0.14)$ $0.155$ $0.079$ $156$ $0.08$ $0.0566$	143	3.81	3.85	3.87	3.84
145 $2.94$ $2.94$ $146$ $2.47$ $2.47$ $147$ $1.70$ $1.65$ $148$ $1.37$ $1.37$ $149$ $0.548$ $0.548$ $150$ $0.314$ $0.314$ $151$ $0.204$ $0.204$ $152$ $0.153$ $0.153$ $153$ $0.242$ $0.242$ $154$ $0.096$ $(0.14)$ $0.155$ $0.724$ $(0.085)$ $0.096$ $0.079$ $156$ $0.08$ $0.0566$	144	5.20	3.52	3.09	3.27
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	145		2.94		2.94
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	140		2.4/	1.07	2.47
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	141		1.70	1.65	1.07
$ \begin{array}{c ccccccccccccccccccccccccccccccccccc$	140		0 649		1.3/
$ \begin{array}{c ccccccccccccccccccccccccccccccccccc$	150		0.340		0.546
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	151		0.204		0.004
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	150		0 157		0.157
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	102		0.100		0.100
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	100		V•242 0.00€	(0,14)	0.242
156 0.08 0.0566 0.062 0.058	154		0.000 0.701	(0, 14)	0.112
	156	0.08	0.0566	(0,062)	0.019
157 0.0429 (0.036) 0.039	157	0.00	0.0429	(0.036)	0.039
			~, ~ i <b>~ v</b>	100001	

TABLE 3 (cont'd)

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Mass No.	von Gunten	Meek & Rider	Crouch	Mean
158 159 160 161 162 163 164 165 166 167 168 169 170		0.0261 0.0176 0.0129 0.0067 0.0031 0.0018 0.0013 0.00061 0.00029 0.00021 0.00012 0.00013 0.00013	(0.022) 0.0127 (0.0074) 0.0051 0.0028 0.0008	0.024 0.015 0.010 0.0059 0.0031 0.0018 0.0013 0.00061 0.00029 0.00021 0.00012 0.00010 0.00010

TABLE 3 (cont'd)

Note: Yields in brackets are interpolated by the evaluator.

# TABLE 4

# Recommended 14 MeV fission yields for ²³⁸U (per cent)

f	ومستهيد بالمراكل والمترا بالمستجوب المتراكي والمراجب				
67		0.00014	0.00014	0.00014	0.00014
68		0.00029		(0.00027)	0.00028
69		0.00049		(0.00048)	0.00048
70		0.00088		(0.00088)	0.00088
71		0.00157	0.000	(0.0016)	0.00158
72		0.00295	0.003	0.0030	0.00298
73		0.00472	0.005	0.0054	0.00504
14		0.00787		(0.0082)	0.00803
75		0.0138		(0.013)	0.00134
76		0.0216	0,0000	(0.019)	0.0203
		0.0354	0.0298	0.030	0.0317
78		0.0570	0.0412	0.041	0.169
79 90		0.100	0.19	(0.18)	0.100
83		0.211	0 74	(0.23)	0.23
82		0.015	0.04	(0.48)	0.46
83	0.65	0.650	0.722	0.683	0.676
84	1,20	1.17	1.315	1,22	1.23
85	1.20	1.01	1.12	1-08	1.07
86		1.55	1.76	1.70	1.67
87		1.85		1.61	1.73
88		2.23		1.75	1.99
89	2.76	2.85	2.70	2.61	2.73
90	3.25	3.17	3.13	2.97	3.13
91	3.35	3.23	3.14	3.55	3.32
92		3.96		4,03	4.00
93	4.30	4.11	4.14	4.41	4.24
94		4.83		(4.75)	4.79
95	5.10	5.10	5.31	5.11	5.15
96		5.39		(5.23)	5.31
97	5.38	5.32	5.49	5.36	5.39
98		5.58		(5.52)	5.55
99	5°95	5.74	5.81	5.69	5.79
100		5.44		(5.79)	5.61
	5.90	6.09	6.02	5.90	5.98
102	3.65	3.78	3.54	(5.12)	4.02
103	3.00	4.49	4.16	4.43	4.02
104		3.82	0.05	(3.62)	3.72
105	3.23	3.36	2.69	2.97	3.06
105	2.40	2.13	2.40	2.14	2.21
		1.11	1.78	1.14	1.70

. .....

$ \begin{array}{ c c c c c c c c c c c c c c c c c c c$	Mass No.	von Gunten	Meek & Rider	Crouch	Daroczy et al	Mean
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	108		1.35		(1.53)	1.44
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	109	1.20	1.42	1.52	1.34	1.37
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	110		0,997		(1.12)	1.06
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	111	0.89	1.10	0.917	0.953	0.965
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$	112	0.70	1,24	0.993	0.985	0.980
$ \begin{array}{c ccccccccccccccccccccccccccccccccccc$	113	0.81	0.973	0.877	0.877	0.884
$ \begin{array}{c ccccccccccccccccccccccccccccccccccc$	114		0.685		(0.85)	0.767
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	115	0.78	0.879	0.741	0.814	0.803
$ \begin{array}{c ccccccccccccccccccccccccccccccccccc$	116		0.675		(0.82)	0.747
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	117		0.845		(0.83)	0.837
$ \begin{array}{c ccccccccccccccccccccccccccccccccccc$	118		0.667		(0.83)	0.748
$ \begin{array}{c ccccccccccccccccccccccccccccccccccc$	119	ļ	0.675	ł	(0.84)	0.757
$ \begin{array}{c ccccccccccccccccccccccccccccccccccc$	120		0,702		(0.84)	0.771
$ \begin{array}{c ccccccccccccccccccccccccccccccccccc$	121	0.81	1,17	0.97	0.851	0.950
$ \begin{array}{c ccccccccccccccccccccccccccccccccccc$	127		0, 781		(0.85)	0.780
$ \begin{array}{c ccccccccccccccccccccccccccccccccccc$	123		0.850	ł	(0.84)	0.845
$ \begin{array}{c ccccccccccccccccccccccccccccccccccc$	120		0.060	1	(0.84)	0.904
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	125	0.70	1 37		0.839	0.970
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	120	0.10	1.26		(1.08)	1 17
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	120	1 52	1.51	1 19	1 40	1.46
$ \begin{array}{c ccccccccccccccccccccccccccccccccccc$	121	1.52	1.01	1.46	(1.92)	2.03
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	120	1 70	2.23	1 26	1 00	1 40
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	129	1.50	2.30	1.20	(3.00)	7 75
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	170	A 77	4 04	7 97		4.03
	170	4.37	4.04	3.05	J. 30	4.03
	177	4.00	4.79	4.00	5.06	6 70
133 $0.02$ $0.10$ $0.04$ $3.50$ $0.00$	133	0.02	6 44	0.04	6.40	6.02
104 4.7 0.44 0.55 0.40 0.02 177 555 565 569	104	4.1	5.07	0.00 5 65	5 65	5.69
	135	3.00	0.01	5.03	5.05	1 16
100 $5.43$ $5.04$ $5.57$ $4.10$ $177$ $6.6$ $5.47$ $70$ $70$ $766$	100	<b>6</b> C	5.45	5.04	3.37	4.10
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	179	0.0	3.47	1 71	4.12	3.00
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	130	A 64	A 01	5.00	4.15	4.03
109 $4.04$ $4.51$ $3.00$ $4.00$ $4.01$	139	4.04	4.51	1.67	-4.00 A 46	4.01
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	140	4.00	4.30	4.07	4.40	4.02
141 3.0 4.33 4.11 4.22 4.11 142 7.01 4.20 4.05	141	3.0	7 01	( 'to / /	4 20	4.77
142 3.51 4.20 4.00	142	3 67	3.31	3 79	3-86	3.75
140 $3.01$ $3.11$ $3.12$ $3.00$ $3.10$ $144$ $3.20$ $7.40$ $3.21$ $7.04$ $3.23$	143	3.07	3 40	3 21	3.00	3.73
	145	0.20	2 94	7 19	7 19	3.04
	146		2.04	0.10	(258)	2 12
147 2.0 2.28 2.10 2.15 2.13	147	2.0	2.20	2 10	(2eJ0) 2.15	2.13
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	148	2.0	1.78	2.10	(1 62)	1.70
149 $132$ $(102)$ $100$	140	}	1 32		(1.02)	1.27
	150		1.13		(0.93)	1.03
	151		0.849		(0.71)	0,779
	150		0 602		(0.54)	0.580
	152	0.70	0 402	0.40	0.408	0.400
	100	0.09	0.400	0.40	(0.97)	0.261
	104		0.156		(0,27) (0,10)	0 169
	100	0.17	0.100	0.11	0.10	0.127
	100		0,100	<b>U</b> .11	V#12	

TABLE 4 (cont'd)

Mass No.	Von Gunten	Meek & Rider	Crouch	Daróczy et al	Mean
157		0.0825		(0.072)	0.0772
158		0.0427		(0.043)	0.0428
159		0.0258	0.026	0.026	0.0259
160		0.0164		(0.0145)	0.0155
161		0,0089	0.0083	0.0085	0.0086
162		0.0059		(0.0051)	0,0055
163		0.0034		(0.0029)	0.0031
164		0.0020		(0.0018)	0.0019
165		0.0011		(0.0010)	0.0010
166		0.00085	0.00063	0.00063	0,00070
167		.0 _* 00037		(0.00037)	0.0003
168		0.00020		(0.00022)	0,0002
169		0.00013	0,00013	0.00013	0.00013

TABLE 4 (cont'd)

Note: Yields in brackets are interpolated by the evaluator.

#### TABLE 5

Recommended 14 MeV fission yields for 237 Np (per cent)

Mass No.	Crouch*
91939799105109111112127131132139140143147153157	2.71 4.49 5.43 4.94 3.50 1.48 1.23 1.23 2.52 3.55 4.29 4.84 4.89 3.60 1.73 0.32 0.094

*Crouch's is the only current evaluation of this nuclide.

#### TABLE 6

Recommended 14 MeV fission yields for ²³⁹Pu (per cent)

Mass No.	Crouch*
89	1.72
91	2.04
97	3.99
99	4.16
103	6.25
106	4.16
111	1.55
113	1.09
115	1.30
132	4.58
137	5.10
140	2.86
144	2.17
147	1.41
156	0.02
161	0.000138

*Crouch's is the only current evaluation of this nuclide.

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238_U fast fission yield data listed by M. Lammer⁽⁷¹⁾

	والمراجع والمراجع والمراجع والمراجع والمترافع	ومقور والمراجعة ومراجعة أوروه والالا الجزاري والإرتبال	موذعين ومغاولة الروسية وال			a a change and the second second second second second second second second second second second second second s	
Fission Product	yield	Standard	Ref.	Fission Product	yield	Standard	Ref.
Sr-89	3,12-0.19	absolute	Cun72	Cd-115g	°033 ⁺ °005	absolute	Cun72
	4.4-0.4	absolute	Bon60		.046007	absolute	Bon60
	3.4-0.3	Mo-99	Pet60		.039004	Mo-99	Pet60
	2.8-0.3	Ba-140	Ke154		<u> </u>	Ba-140	Ke154
	3.2	Ba-140	Eng52	I-131	3.62-0.11	absolute	Lar72
Zr-95	7.24-1.31	absolute	Cun72	<u>Xe-131</u>	2.16	Ba-140	Mat72
	5.47-0.19	absolute	Lar72	Te-132	4.23-0.34	abso lute	Cun72
	5.2-0.6	absolute	Bon60		5.27-0.32	absolute	Lar72
	6.1 [±] 0.6	Mo-99	Pet60		4.1-0.4	absolute	Bon60
	4.9-0.7	Ba-140	Ke154		4.9-0.6	Ba-140	Ke154
	7.1	Ba-140	Eng52	Xe-132	4.64	Ba-140	Mat72
Zr-97	6.00+0.37	absolute	Cun72		5.31	I-131	Mat72
1	5.91-0.18	absolute	Lar72	Cs-137	7.68-1.72	absolute	Cun72
	5.2-0.6	absolute	Bon60		6.1-0.7	absolute	Bon6O
Mo-99	6.00+0.79	absolute	Cun72		7.4-0.7	Ba-140	Ke154
	6,14-0,18	absolute	Lar72		5.15	Ba-140	Mat72
	7.0-0.7	absolute	Bon60		5.88	I-131	Mat72
	6.6+0.4	absolute	Pet60		6.52	Nd	Rid67
	6.7-0.7	Ba-140	Ke154	Ba-140	6.03-0.42	absolute	Cun72
	5.74	Ba-140	Eng52		5.96-0.17	bsolute	Lar72
Ru-103	6.26-0.19	absolute	Lar72		$5.8^{+}_{-}0.5$	absolute	Bon60
	3.9-0.5	absolute	Bon6O		6.7-0.5	absolute	Pet60
	6.6-1.0	Ba-140	Kel54		6.03-0.19	U-235	Ciu68
	7.2	<u>Ba-140</u>	Erg52		5.72-0.14	U-2 <b>35</b>	Die71
Ru106	2.85-0.30	absolute	Bon60		(6.79)	I-131	Mat 72
	3,02+0,30	Ba-140	Ke154	Nd-143	4.42	Ba-140	Mat72
	2.63	<u>Ba-140</u>	Eng52		4.59	Nđ	Rid67
Ag-111	.058 ⁺ .011	absolute	Cun72	Ce-144	3.55	R-value	Bun58
	<b>.</b> 094 ⁺ <b>.</b> 012	absolute	Bon60	and	3.91	R-value	Bun58
	.087008	Mo-99	Pet60	Nd-144	5.1-0.5	Ba-140	Ke154
	.067 ⁺ .006	Ba-140	Ke154		4.56	Ba140	Mat72
	.070	Ba-140	Eng52		4.53	Nd	Rid67

APPENDIX 3 (cont'd)

Fission Product	yield	Standard	Ref.	Fission Product	yield	Standa <b>rd</b>	Ref.
Nd-145	3.72	Ba-140	Mat72	Nd-150	1.27	Ba-140	Cun72
	3.69	Nd	Rid67		0.90	Nd	Rid67
Nd-146	3.36	Ba-140	Mat72	Eu-156	0.042+0.008	absolute	Cun72
	3.33	Nd	Rid67		0.072	R-value	Bun58
Nd-148	2.08	Ba-140	Mat72		0.065	R-value	Bun58
	2.03	Nd	<u>Rid67</u>		0.076-0.010	Ba-140	Ke154
					0.061	Ba-140	Eng52

#### Standard:

absolute: number of fissions determined for each sample.

- Mo-99: renormalized to Mo-99 yield of 6.14 (Lar72).
- Ba-140: renormalized to Ba-140 yield of 5.94 (unweighted average of Cun72, Lar72, Ciu68 and Die71).
- I-131: renormalized to I-131 yield of 3.62 (Lar72).
- Nd: sum of Nd yields without Nd-150 of Rid67 normalized to the sum of the same yields of Mat72 (rel Ba-140).
- R-value: yields rel Mo-99 as ratio to U-235 thermal yields.
- U-235: measured relative U-235 thermal fission yield of Ba-140 (6.36% used here).

#### References

In addition to the ref code used here, the ref number used in the UKAEA fission product library (E...C. Crouch, AERE R 7394 (1973) is also given.

166	Bon60	E.K. Bonyushkin et al, AEC-tr-4682(1960)
37	Bun58	L.R. Bunney et al, 2nd Int. Conf. PUAE, Geneva 1958,
		Vol. 15, p. 449.
157	Ciu68	L. Ciufollotti, Energia Nucleare 15(1968) 272
634	Cun72	J.G. Cuninghame et al, AERE-6862(Rev)(1972)
608	Die71	R. Dierckx et al, J. Nucl. En. <u>25(1971)</u> 85
179	Eng52	D.W. Engelkemeir et al, ANL-4927(1952)
234	Ke154	R.N. Keller et al, Phys. Rev. <u>94(1954)</u> 969
	Lar72	R.P. Larsen et al, Trans. Am. Nucl. Soc. 15(1972) 483
	Mat72	C.K. Mathews et al, Can. J. Phys. <u>50(1972)</u> 3100
2	Pet60	K.A. Petrzhak et al, AEC-tr-4696(1960)
	Rid67	B.F. Rider et al, GEAP-5505(1967) 21.





THE ONLY CURRENT EVALUATION (CROUCH), WHOSE VALUES ARE GIVEN IN APPENDIX I, TABLE 2. THE CIRCLES SHOW SIDEBOTHAM'S CALCULATED VALUES.





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FIG II. EXPERIMENTAL FAST FISSION YIELDS FOR 235U IN EBR-II : NEW RESULTS PRODUCED BY W. J. MAECK FOR THIS REVIEW PAPER.



THE MEAN VALUES GIVEN IN APPENDIX 2, TABLE 1.



FIG. 13. 14 MeV FISSION YIELDS FOR 2001: THE BLACK CIRCLES ARE THE VALUES FROM THE ONLY CURRENT EVALUATION (CROUCH), GIVEN IN APPENDIX 2, TABLE 2.



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THROUGH THE MEAN VALUES GIVEN IN APPENDIX 2, TABLE 3.





VALUES FROM THE ONLY CURRENT EVALUATION (CROUCH), GIVEN IN APPENDIX 2, TABLE 5.














FIG. 23.



FIG. 24. ALL AVAILABLE FRACTIONAL CHAIN YIELDS FOR FAST AND 14 MeV FISSION PLOTTED AS A FUNCTION OF Z-ZP.

