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STATUS AND INFORMATION NEEDS

M. Lammer
Nuclear Data Section, IAEA, Vienna

(Paper presented at the IAEA Consultants' Meeting on

Evaluation of the Quality of Safeguards Neutron Coincidence Measurements,

Vienna, 24-28 November 1986)

December 1986

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1. INTRODUCTION

The Nuclear Data Section (NDS) of the IAEA is preparing the publication of a Handbook on Nuclear Data for Safeguards. This handbook will be the result of a cooperation of NDS with safeguards experts to define the data needs, and with nuclear data experts for recommending the best available data. NDS will only extract data from existing evaluations and include only data in the handbook for which recommendations from data experts are being received.

Some problems are still encountered in the preparation of a complete handbook, as requested by safeguards experts:

- for some data the way of their representation in the handbook has still to be decided; (to be answered by safeguards experts)
- other data are presently not available, or their availability is not known to NDS (to be answered by data experts).

These two problem areas are essentially the "information needs".

In order to make the existing data available to safeguards users as soon as possible, a first version of the handbook will be produced by the end of this year. A draft will be sent to data experts for review. The first version will contain the most important and readily available data. Evaluation of some more data is presently under way. These data will be included in the handbook next year, possibly together with further existing nuclear data, for which recommendations may be received.

In order to facilitate amendments and updates to the first version, the handbook will be distributed in loose-leaf format: A further advantage of this format is that safeguards' users can easily take out those tables from the handbook which they need for their own field of application. Recipients of the handbook will automatically receive updates and amendments.

This paper reviews the data requirements as presented by safeguards experts and the status of data which will be included in the first version of the handbook, and concludes with a list of open questions and problems encountered when all the requested information is to be included in the handbook.

Unfortunately, only a list of nuclear and other data but not their accuracies needed to satisfy safeguards requirements were received. Therefore a comparison of the data status (i.e.: the accuracies achieved) with the requested accuracies is not possible. In this context, I want to draw the attention of safeguards users to WRENDA [1], the "World Request List of Nuclear Data", published by NDS. WRENDA offers the opportunity for safeguards users to publish requests for nuclear data measurements or evaluations, if the data are not available or not sufficiently accurate. WRENDA receives world-wide response from measurers and evaluators.

2. SAFEGUARDS REQUIREMENTS

A questionnaire was sent to safeguards experts, asking them to formulate their nuclear data requirements. Their replies are summarized below, sorted by data type. An extract from the evaluation of the questionnaire containing specific questions and collected replies as well as data requirements sorted by safeguards method are reproduced in Appendix A.

The following abbreviations are used in the tables:

b	barn
d	days
dn	delayed neutron data (Pn, yields, time dependent energy spectra)
$d\gamma$	delayed gamma-ray yields and time dependent energy spectra
E(n)	energy spectrum of prompt neutrons
E_x	energy of radiation x
fast	fission neutron spectrum (specifically U-235 or Cf-252)
I	reduced resonance integral
M(n)	multiplicity distribution of prompt neutrons
$M(\gamma)$	multiplicity distribution of prompt gammas
T1/2	total half-life
T_{sf}	partial half-life for spontaneous fission
T_α	partial half-life for α-decay
X-rays	(in table headings) energies and intensities of X-rays
y	years
$Y(\gamma)$	prompt γ-ray yields and correlations in fission
α-rays	α particle energies and intensities
γ-rays	(in table headings) energies and intensities of γ-rays
ν_p	average number of prompt neutrons emitted per fission
σ_c	thermal reactor spectrum averaged neutron capture cross-section
σ_f	thermal reactor spectrum averaged neutron fission cross-section
$\sigma_c(E)$	neutron capture cross section } as function of neutron energy
$\sigma_f(E)$	neutron fission cross section } as function of neutron energy

2.1. Actinides (Table 1)

The following data will be included in the first version of the handbook:

- total and partial half lives
- γ-ray and α-ray energies and intensities
- thermal cross-section data and resonance integrals.

Energy dependent cross section data are available from computer files upon request. They constitute a large number of individual data points, and the question of their inclusion in the handbook as tables is still open.

The preparation of X-ray, prompt neutron and delayed neutron data is in progress, and amendments to the handbook can be expected for 1987.

2.2. Fission products (Table 2)

All requested data will be included in the handbook.

Table 1: Requested nuclear data for actinides

Actinides decay into long- and short lived daughter products, some of which will also be present in nuclear fuel. The following daughter products will be included in the handbook:

Hg-206; Tl-206 to 210; Pb-205,209 to 212,214; Bi-210 to 215; Po-209 to 216,218; At-215,217 to 219; Rn-217 to 220,222; Fr-221,223; Ra-223 to 226,228; Ac-225,227,228; Th-227 to 230.

The following data types will be given in addition to those listed:

- decay branching fractions, if more than one decay mode is to be considered.
- neutron capture branching to isomeric states, if relevant.

nuclide	T1/2	T_α	T_{sf}	X-rays	α-rays	γ-rays	σ_f	σ_c	σ_f(E)	σ_c(E)	ν_p	M(n)	E(n)	M(γ)	Y(γ)	dn	dγ
Th-231	+			+	+												
Th-232	+	+	+	+	+	+	fast	+	+	+	+	+	+	+	+	+	+
Th-234	+			+	+												
Pa-231	+			+	+												
Pa-233	+			+	+												
Pa-234g,m	+			+	+												
U-232	+	+		+	+	+											
U-233	+	+	+	+	+	+	+	+	+	+	+	+	+	+	+	+	+
U-234	+	+	+	+	+	+		+									
U-235	+	+	+	+	+	+	+	+	+	+	+	+	+	+	+	+	+
U-236	+	+	+	+	+	+		+			+						
U-237	+			+	+		+	+									
U-238	+	+	+	+	+	+	fast	+	+	+	+	+	+	+	+	+	+
Np-237	+	+	+		+	+	+	+									
Np-239	+			+	+			+									
Pu-236	+	+				+											
Pu-238	+	+	+	+	+	+	+	+	+	+	+	+	+	+	+	+	+
Pu-239	+	+	+	+	+	+	+	+	+	+	+	+	+	+	+	+	+
Pu-240	+	+	+	+	+	+	fast	+	+	+	+	+	+	+	+	+	+
Pu-241	+	+	+	+	+	+	+	+	+	+	+	+	+	+	+	+	+
Pu-242	+	+	+	+	+	+	fast	+	+	+	+	+	+	+	+	+	+
Am-241	+	+	+	+	+	+	+	+	+	+	+	+	+	+	+	+	+
Am-242g,m	+		+				+	+	+	+							
Am-243	+	+	+	+		+	+	+									
Cm-242	+	+	+	+		+	+	+			+	+	+	+	+		
Cm-243	+	+	+	+		+	+	+			+	+	+	+	+		
Cm-244	+	+	+	+	+	+	+	+			+	+	+	+	+	+	+
Cf-252	+	+	+	+	+	+					+	+	+	+	+	+	+

ν_p, M(n), E(n), M(γ) and Y(γ) will be given for neutron induced (thermal, fast for selected nuclides) fission and also for spontaneous fission of some nuclides.

Table 2: Requested fission product nuclear data:

The handbook will include:

- complete sets of chain yields and selected cumulative yields from thermal neutron fission of U-233,235, Pu-239,241, and from fast neutron fission of U-238,
- decay and cross section data as indicated in the table below,
- branching fractions to isomeric states in decay and neutron capture.

nuclide	T1/2	γ-rays	σ_c	nuclide	T1/2	γ-rays	σ_c
Kr [1])			+	Ba-140	+	+	
Kr-85	+		+	La-140	+	+	
Zr [1])			+	Ce-141	+	+	
Zr-95	+	+		Ce-144	+	+	+
Nb-95m	+	+		Pr-141			+
Nb-95	+	+		Pr-144	+	+	
Mo [1])			+	Nd [1])			+
Ru [1])			+	Nd-147	+	+	+
Ru-103	+	+		Pm-147	+		+
Ru-106	+			Pm-148m	+		+
Rh-106		+		Pm-148	+		+
Sb-125	+	+		Pm-149	+		+
I-131	+	+		Pm-151	+		+
Xe [1])			+	Sm [1])	+		+
Xe-133	+		+	Sm-151	+		+
Xe-135	+		+	Sm-153	+		+
Cs-133			+	Eu-153			+
Cs-134	+	+	+	Eu-154	+	+	+
Cs-137	+	+	+	Eu-155	+	+	+

[1]) Stable isotopes for mass-spectrometric measurements

Fission products with A=147-153 are included for calculating the formation of Eu-154

The fission products Ru-103, Sb-125, I-131, Ce-141 and Eu-155 are included as their γ-rays may interfere with those of other fission products (depending on cooling time).

2.3. Reactions with light nuclei:

(n,x) reaction cross-sections with x = charged particles relevant for neutron counters for the following nuclei:

H, D, He-3,4, Li-6, Be-9, B-10, Cd-113

(α,n) cross-sections vs. Eα (reaction threshold below Eα ≃ 7 MeV) for:

Li, Li-6,7, Be-9, B, B-10,11, C, C-13, N, O, O-17,18, F-19, Na-23, Mg, Al-27, Si, P-31, S, Cl, K, Ca.

(γ,n) cross-sections for Be-9, D_2O and other materials still to be specified

No recommendations for such data were received yet. These data do exist in handbooks and on files, and will probably be included in the second issue of the handbook.

2.4. Non-neutron and macroscopic data, additional presentations

a) γ-ray attenuation coefficients as function of γ-ray energy for different nuclear fuel, construction and shielding materials (still to be defined in detail);

b) Spontaneous fission rates and neutron yields per gram of isotope.

c) (α,n) yields per gram of compound for oxides, carbides, fluorides and alloys.

d) Thick target (α,n) yields per α vs. α-energy for C,O,F and Al.

e) Same as above, but with α-spectra from the α-decay of the isotopes of U, Pu etc.

f) Data for isotopic neutron sources (yield/g, yield/Ci, energy spectra, average energy, etc).

g) Photo-absorption cross sections, fluorescent yields for fissile and fertile materials.

h) "Twin-target yields" for UF_6.

i) Specific heat generation rates (watts/gram) fro the Pu and Am isotopes.

j) Physical and Chemical data for typical nuclear material (density, stöchiometric data, solubility etc. of U,Pu,UO_2,PuO_2 etc.)

k) Efficiency for typical γ-detectors.

l) Graphical presentations of gamma spectra from typical fresh and spent fuel.

m) (possibly) list of gamma rays sorted by increasing energy.

n) natural isotopic abundance of U and fission products (table 2).

For a), references will be given in the handbook. The list of γ-rays sorted by increasing energy (item m) can easily be extracted and produced from the tables of γ-rays sorted by nuclide.

In addition to the data suggested above, the inclusion of the following items was requested in individual replies:

- References to more detailed data and information than given in the handbook.
- Models and approximation formulae for:
  - neutron cross section data
  - fission neutron energy distributions
  - multiplicity distributions
  - time dependence of delayed neutron and gamma-ray spectra
  - burnup calculation

- Graphical presentations of
  - excitation functions (e.g. for (α,n), (γ,n) etc. reactions),
  - spectra of fission neutrons and fission gammas,
  - spectra of delayed neutrons and gammas (time dependent)
  - spectra of γ-ray standard sources

- α- and γ-ray calibration data
- Selected physical and fundamental constants
- Intrinsic line widths in X-ray spectra.

Since these suggestions were not supported by other replies, I want to ask safeguards experts: is the inclusion of such information in the handbook desirable; if yes, which information? For some items: which sources, constants etc.?

3. DATA STATUS

The following tables present the status of those nuclear data which I have received already for inclusion in the first version of the handbook. All uncertainties are given as 1 standard deviation.

3.1. Actinides

Decay data and their status (Tables 3-5) are taken from the final report [2] of the IAEA Coordinated Research Programme on the measurement and evaluation of transactinium isotope nuclear decay data, which was carried out from 1978 until 1985.

Table 3-a shows that accuracies of total half-lives for the most important actinides are better than 0.5%, with the exception of Pu-241: The reason for existing descrepancies between earlier measurements is still not fully understood and resolved. Only the inclusion of recent, more consistent measurements brought the uncertainty down to 0.7%, which is higher than that of individual measurements, and can probably only be reduced by resolving the discrepancies.

Uncertainties of some partial spontaneous fission half life values collected in Table 3-b are rather high. On the other hand, the branching fractions for spontaneous fission are several orders of magnitude smaller than the total half lives. They may, however, be still important for passive neutron assay in safeguards. Safeguards experts should decide whether the present accuracies for spontaneous fission half lives, and also for the α-particle data presented in Table 4, are sufficient.

Table 3-a: Actinides: Status of total half-lives

range of relative uncertainties (%)	nuclides (in some cases the relative uncertainty [%] is given in brackets)
> 1	Pa-234m (2.6), Pu-236 (3), Am-242m (1.4)
0.5 - 1	Pa-234 (0.75), U-232 (0.7), Np-237 (0.5), Pu-241 (0.7), Cm-243 (0.7)
0.2 - 0.5	Th-232, Pa-231, Pa-233, Pu-238, Pu-242, Am-243 (0.2), Cf-252
0.1 - 0.2	Th-234, U-233 through U-238, Np-239, Pu-239, Pu-240, Am-241, Am-242, Cm-244
< 0.1	Th-231, Cm-242

Table 3-b: Actinides: Status of spontaneous fission half-lives

nuclide	relative uncertainty (%) 1)	branching fraction 2)	nuclide	relative uncertainty (%) 1)	branching fraction 2)
Th-232	n.u.	E-11	Pu-241	n.a.	
U-233	n.u.	E-13	Pu-242	1.5	E-6
U-234	5.6	E-11	Am-241	2.1	E-12
U-235	29	E-11	Am-242	n.a.	
U-236	2.9	E-10	Am-242m	36	E-10
U-238	1.2	E-7	Am-243	25	E-11
Np-237	n.u.	E-12	Cm-242	2	E-8
Pu-238	4	E-9	Cm-243	n.a.	
Pu-239	n.u.	E-12	Cm-244	0.5	E-6
Pu-240	3.4	E-8	Cf-252	0.35	0.031

1) n.u. = no uncertainty available; n.a. = no half life value available
2) Generally given as E-x meaning 10^{-x}

Table 4-a: Actinides: status of α-ray energies

Given below are energy uncertainties (u) exceeding 1 keV for α-rays with intensities > 1% of all α-decays.

nuclide	u (keV)	nuclide	u (keV)
Th-232	3-8	Np-237	1.5-2
U-233	1.2	Pu-241	1.2-3
U-235	2-5	Pu-242	1.2
U-236	3-5	Cm-243	1-3
U-238	5		

Table 4-b: Actinides: status of α-ray intensities

Given below are relative uncertainties of α-ray intensities exceeding 10% of all α-decays, and in brackets for 1% < intensity < 10%.

nuclide	uncertainty (%)	nuclide	uncertainty (%) [1]
Th-232	2.5-9	Pu-240	0.7-2
U-232	0.6-1.3	Pu-241	1-1.7 (10-20)
U-233	0.4-1.3 (2.7)	Pu-242	0.2-0.7
U-234	0.03-0.07	Am-241	0.9-1.6 (14)
U-235	5.5-10 (10)	Am-243	n.u.
U-236	5-15	Cm-242	0.7-2
U-238	5-17	Cm-243	1.4-2 (n.u.)
Np-237	19-24 (2-38)	Cm-244	0.07-0.2
Pu-236	1.2-2.8	Cf-252	0.4-2
Pu-238	0.08-0.2		
Pu-239	1-1.7		

[1] n.u. = no uncertainties available

Table 5: Actinides: status of γ-ray intensities

Given below are relative uncertainties of intensities of the most significant γ-rays above 200 keV, and in brackets of those below 200 keV.

nuclide	uncertainty (%)	note	daughter(s)	comment
Ra-224	0.17-4(2.5-5)	a,b	Pb-212,Bi-212,Tl-208	most significant γ-rays from Pb-212,Tl-208
Th-231	1.5-8 (20)	β^-		
Th-232	see Ra-224	b	Ra-228,Th-228,Ra-224	see Ra-224,Th-232 γ-s negligible
Th-234	(20)	β^-		
Pa-231	2.2-3(2.7-4)	c	Ac-227,Th-227,...	significant γ-s from daughter
Pa-233	0.9-1.5	β^-	U-233	
U-232	see Ra-224	b	Th-228, Ra-224,...	see Ra-224,U-232 γ-s negligible
U-233	1.2-4(1.4-2)	b	Th-229,...	significant γ-s from Th-229,Fr-221,Bi-213
U-234	(~1.5)	c	Th-230,Ra-226	intensities: $\sim10^{-5}$% (<200 KeV: 0.1+0.03%)
U-235	1-2(0.7-0.9)	c	Th-231,Pa-231	186 KeV: 0.9%; for daughters see Pa-231
U-236	(10-25)	c,d	Th-232,...,Ra-224,...	intensities: 0.078+0.019%
U-237	~2.5	β^-	Np-237	
U-238	see daughters		Th-234,(Pa-234),U-234	U-238: 50 KeV γ-ray negligible
Np-237	all: 1.1-2.2	c	Pa-237	
Np-239	all ~1.5	β^-	Pu-239	
Pu-238	(1-2)	c,d	U-234,...	intensities about 10^{-2}-10^{-3}%
Pu-239	0.6-2(1-2)	c,d	U-235,...	
Pu-240	(0.8-1.6)	c,d	U-236,Th-232,	
Pu-241	(0.9-2)	β^-	Am-241	
Pu-242	(2.6)	c,d	U-238,...	intensities about 10^{-2}-10^{-4}%
Am-241	2-10 (1-5)	d	Np-237	
Cm-244	(2.4-10)	d	Pu-240	
Cf-252	(>6)			

Notes:
a: daughter product
b: uncertainties of equilibrium data
c: data without daughter products
d: daughter(s) have longer half-life
β: β-decay (no daughters)

Table 6: Actinides: status of neutron cross-section data

type: fission (f) or capture (c) cross section as function of neutron energy ($\sigma(E)$), or as $\hat{\sigma}$ from thermal data and resonance integral (RI)

u: relative uncertainty of reactor spectrum averaged cross section from $\hat{\sigma}$ or $\sigma(E)$

d: relative (average) discrepancies between measurments and/or evaluations

res: yes .... significant contribution from epithermal region
dom .... contribution from epithermal region dominates

nuclide	type	u(%)	d(%)	res	comment
Th-232	$\sigma c(E)$	1-3		yes	u: higher value for higher fraction of epithermal neutron flux
U-233	$\sigma c(E)$	2-3		yes	deduced from other data
	$\sigma f(E)$	0.5-1			
U-234	$\hat{\sigma} c$	2-6	8	yes	RI: u = 11%; d between JENDL-2 and ENDF/B
U-235	$\sigma c(E)$	1-2			
	$\sigma f(E)$	0.2-0.5			
U-236	$\hat{\sigma} c$	~4	7	dom	d between JENDL-2 and ENDF/B at 1eV-1keV
U-237	$\hat{\sigma} c$	30-40			
U-238	$\sigma c(E)$	~1		dom	
Np-237	$\hat{\sigma} c$	2-5	4-14	yes	d: 4%: 1 eV-1 keV, 7-14%: <1 eV
	$\hat{\sigma} f$	15	15-50	dom	d: 15% <1 eV, 50%: 1 eV-1 keV
Pu-238	$\sigma c(E)$	1.5-2	~2		
	$\sigma f(E)$	3-5			high u and d at 1 eV-1 keV only
Pu-239	$\sigma c(E)$	1.5-2			
	$\sigma f(E)$	0.5-1			
Pu-240	$\sigma c(E)$	2-2.5	4.5	dom	d: at 1 eV-1 keV only
Pu-241	$\sigma c(E)$	~1.5	~10		d: 10%: <1 eV, 1% at 0.025 eV
	$\sigma f(E)$	0.6-1	2.2		d: 2.2%<1 eV, 0.7% at 0.025 eV
Pu-242	$\sigma c(E)$	3-4	~4	dom	
Am-241	$\sigma c(E)$	~3	4-5		d: between JENDL-2 and ENDF/B
	$\sigma f(E)$	~4	8-10		d: recent measurement agrees with evaluations
Am-242m	$\hat{\sigma} c$ 1)	30	40		measured: $\hat{\sigma}$ only, d: to res. calc.
	$\sigma f(E)$	~4	(20)		d: in RI (insignificant)
Am-242	$\sigma c(E)$				calculated values only
	$\hat{\sigma} f$ 1)	10			measured: $\hat{\sigma}$ only
Am-243	$\hat{\sigma} c$	3-4?	20	dom	d in RI between measurement and res.calc.
	$\hat{\sigma} f$	10?	high	dom	d: factor 2-4 among measurements and to res. calc.
Cm-242	$\hat{\sigma} c$	25-30		yes	evaluations agree
	$\hat{\sigma} f$	?	~60		uncertainties not evaluated
Cm-243	$\hat{\sigma} c$	8-10	~60		d: between JENDL-2 and ENDF/B
	$\hat{\sigma} f$	4-5	10-15		d: between JENDL-2 and ENDF/B
Cm-244	$\hat{\sigma} c$	5-6	15-30	dom	d: between JENDL-2 and ENDF/B
	$\hat{\sigma} f$	20	50-70		d: between JENDL-2 and ENDF/B

1) For the requested $\sigma(E)$ only data calculated from resonance parameters exist, which are discrepant to measurements of $\hat{\sigma}$. The uncertainty is given for the measured $\hat{\sigma}$.

The handbook, in its first version, will present γ-ray data only for parent nuclide decays. The inclusion of equilibrium decay γ-ray data is not only a question of needs (as discussed further below), but also only few of those as data are available. Among those listed in Table 5 (note b) as equilibrium decay γ-data, only those of U-232 (daughter: Ra-224) are directly available; for the others, the uncertainties presented in the table have been deduced from those given for the daughters listed.

Thermal cross section data for the handbook and the uncertainties in Table 6 ($\hat{\sigma}$) are taken from the BNL evaluation [3], the status of $\sigma(E)$ data from recent reviews [4], where uncertainties are given for different incident neutron energy ranges.

The uncertainties given in Table 6 are those of thermal reactor-spectrum averaged cross-sections calculated for:

- $\hat{\sigma}$ according to the Westcott formalism [5]

$$\hat{\sigma} = \sigma_o(g+rs) = g\sigma_o + r\sqrt{4/\pi}\sqrt{T/T_o}.I$$

where σ_o = 2200 m/s cross section
g = factor describing the deviation of $\sigma(E)$ from 1/v behaviour
r = epithermal index
T = Maxwellian neutron temperature
T_o = 293° K
I = reduced resonance integral (1/v part subtracted)

- $\sigma(E)$ from point or group cross sections or resonance parameters, folded over a thermal reactor neutron spectrum.

Typical values for the epithermal (1/E) flux, ϕe, relative to the thermal (Maxwellian) flux, ϕm, in the range

$$r \approx \phi e/\phi m = 0.05 - 0.2$$

(exact: $r = \sqrt{\pi/4}\,\phi e/\phi m$, with $\phi m = nv_o\sqrt{T/T_o}$; for conversions see, e.g. [6])

were used for deriving the overall uncertainties given in Table 6.

There are only few cross-sections with relatively high uncertainties. Especially those of Am and Cm isotopes may be significant for the calculation of Cm buildup and neutron emission rates (neutron assay techniques).

The table shows that, for some $\sigma(E)$ data the discrepancies between evaluations is larger than the uncertainties claimed therein.

3.2. Fission products

Fission product decay data are extracted from the Berkeley file which forms the data base for the "Table of Radioactive Isotopes" [7].

Half lives are quite accurately known, with two important exceptions:

- Cs-137: discrepancies among experimental data, ranging from 29.4 to 30.7 years and claiming overall errors of 0.2 y and less, are still not resolved.

Table 7-a: Fission products: status of half-lives

range of relative uncertainties (%)	Nuclides (for the most important fission products the uncertainty [%] is given in brackets)
> 0.5	Nb-95m, Sb-125, Cs-137 (0.67), Sm-151 (7), Eu-154 (1.1)
0.1-0.5	Kr-85, Ru-106 (0.29), Xe-133, Cs-134 (0.24), Xe-135 (0.22), Pm-148m, Pm-151, Sm-153, Eu-155
< 0.1	Zr-95 (0.06), Nb-95, Ru-103, I-131, Ba-140, La-140, Ce-141, Ce-144 (0.07), Nd-147, Pm-147

Table 7-b: Fission products: selected half life values

Zr-95:	64.02 ± 0.04 d	Cs-137:	30.0 ± 0.2 y
Nb-95:	34.97 ± 0.03 d	Ce-144:	284.9 ± 0.2 d
Ru-106:	1.020 ± 0.003 y	Eu-154:	8.8 ± 0.1 y
Cs-134:	2.062 ± 0.005 y		

Table 8: Fission products: status of γ-ray intensities

Uncertainties (u) of relative intensities of the more important fission products

nuclide	u(%)	nuclide	u(%)	nuclide	u(%)
Zr-95	0.4-2	Cs-134	0.3-2	Ce-141	0.8 [1)]
Nb-95	<0.1 [1)]	Cs-137	0.08 [1)]	Ce-144	3.6 [1)]
Ru-103	2.6-3	Ba-140	1-2	Pr-144	1.0-3.8
Rh-106	3-5	La-140	<0.1-2	Eu-154	6 [2)]

1) One γ-ray

2) main contribution: systematic uncertainty of 5.6%

Table 9: Fission product cross section data

nuclide	σ_o(b)	uncertainty b	uncertainty %	I (b)	uncertainty b	uncertainty %	notes
Kr-83	180.	30.	17	183.	25.	14	
Kr-85	1.66	0.2	12	1.8	1.0	8.5	
Zr-91	1.24	0.25	20	5.2	0.7	13	
Zr-93	$1.3<\sigma<4$						
Mo-95	14.0	0.5	3.6	109.	5.	5	
Mo-97	2.1	0.5	25	14.	3.	20	
Ru-101	3.4	0.9	26	100.	20.	20	
Ru-102	1.21	0.07	6	4.2	0.1	2.4	
Xe-131	85.	10.	12	900.	100.	11	
Xe-133	190.	90.	47				a
Xe-135	2 650 000.	110 000.	4.2	7 600.	500.	7	
Cs-133	29.	1.5	5	437.	26.	6	
Cs-134	140.	12.	8.6	(54.)			a
Ce-144	1.0	0.1	10	2.6	0.3	10	
Pr-141	11.5	0.3	3	17.4	2.0	22	
Nd-143	325.	10.	3	128.	30.	25	
Nd-144	3.6	0.3	8	3.9	0.5	13	
Nd-145	42.	2.	5	240.	35.	15	
Nd-146	1.4	0.1	7	3.2	0.5	15	
Nd-147	440.	150.	34	(405.)			
Nd-148	2.5	0.2	8	14.	1.	7	
Nd-150	1.2	0.2	20	14.	2.	15	
Pm-147	168.4	3.5	2	2 064.	100.	5	
Pm-148m	10 600.	2 500.	25	3 600.	2 400.	67	
Pm-148	2 000.	1 000.	50				a
Pm-149	1 400.	300.	20				a
Pm-151	173/<700						a,b
Sm-147	57.	3.	5				
Sm-148	2.4	0.6	25	27.	14.	50	
Sm-149	40 140.	600.	1.5	3 390.			
Sm-150	104.	4.	4	358.	50.	14	
Sm-151	15 200.	300.	2	3 250.	160.	4.5	
Sm-152	206.	6.	3	2 970.	100.	3	
Sm-153	420.	180.	40				
Sm-154	8.4	0.5	6	32.	6.	5	
Eu-153	312.	7.	2.2	1 420.	100.	7	
Eu-154	1 340.	130.	10	802.			
Eu-155	3 950.	125.	3.2	15 300.	2 700.	18	c

The following fission products (not listed above) have cross sections < 1b with relative uncertainties (in %) given in brackets:
Kr-84 (14), Kr-86 (70), Zr-90 (45), Zr-92 (30), Zr-94 (4.8), Zr-96 (4.4), Mo-98 (4.6), Mo-100 (1.5), Ru-104 (6), Ru-106 (30), Xe-132 (13), Xe-134 (7.5), Xe-136 (8), Cs-137 (30)

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notes:

a .... reactor spectrum averaged cross section $\hat{\sigma}$

b .... 1st value: Abgjan [8], 2nd value: BNL [3]

c .... I from Sekine et al [9]; they measured $\sigma_o = 3760 \pm 170$ b

Table 10: Relative uncertainties (%) of cumulative fission product yields

fission product	thermal yields U-233	U-235	Pu-239	Pu-241	fast yields Th-232	U-238
Kr-83	1.0	0.5	0.7	2	2	1.4
Kr-84	1.0	0.7	2	2	2	1.4
Kr-85	3	3	3	3.2	3.2	3
Kr-86	1.4	0.5	0.7	2	2	1.0
Zr-90	1.4	1.0	2	2	4	1.4
Zr-91	1.0	1.0	1.0	2	2	2
Zr-92	1.4	1.0	1.4	2	2.8	2.8
Zr-93	1.4	0.7	1.4	2	4	2
Zr-94	1.4	1.0	2	2	6	4
Zr/Mo-95	1.4	1.0	2	2	2.8	1.0
Zr-96	1.0	1.0	2.8	2	6	4
Mo-97	1.4	0.7	2	2	1.4	0.7
Mo-98	1.4	1.0	2	2	6	1.0
Mo-100	1.4	1.0	4	2	6	1.0
Ru-101	1.4	1.0	1.4	2	11	1.4
Ru-102	1.4	1.0	2	2	11	1.0
Ru-103	2	1.0	2	4	4	1.0
Ru-104	2.8	1.4	2	2	11	1.0
Ru-106	2	1.4	2.8	2	8	1.4
Sb-125	11	4	8	8	11	8
I/Xe-131	1.0	0.5	0.5	1.4	2	1.4
Xe-132	1.4	0.35	0.7	1.4	1.4	1.4
Xe/Cs-133	1.0	0.35	0.7	1.0	2	0.5
Xe-134	1.0	0.5	0.7	1.4	2	2
Xe-135	2	0.35	0.7	1.4	2	0.7
Xe-136	4	0.35	1.0	1.4	2	2.8
Cs-137	0.7	0.5	0.5	1.4	4	0.7
Ba-140	1.4	0.7	1.0	2	2	0.7
Ce/Pr-141	2.8	1.0	2	1.4	2.8	2
Nd-143	1.0	0.35	0.5	1.4	2	0.7
Ce/Nd-144	1.0	0.35	0.5	1.0	2.8	0.7
Nd-145	1.0	0.35	0.5	1.0	2.8	0.7
Nd-146	0.7	0.35	0.5	1.4	2.8	0.7
Nd/Pm-147	2.8	0.5	1.4	1.4	4	1.0
Nd-148	1.0	0.35	0.5	1.0	2.8	0.7
Sm-149	2.8	1.0	1.0	1.4	6	1.0
Nd-150	1.4	0.5	0.5	1.4	16	1.0
Pm/Sm-151	2	0.7	2	1.4	6	1.4
Sm-152	2.8	1.0	1.4	1.4	16	1.0
Sm/Eu-153	6	2.8	6	4	8	2
Sm-154	2.8	1.0	1.4	2	23	1.0
Eu-155	23	4	11	8	23	16

- Eu-154: the adopted value is derived from recent measurements (after 1970) yielding results between 8 and 9 y, which are in conflict with 2 earlier measurements (1952, 1953) of about 16 y. Two independent comparisons of calculated Eu-154 activities with measured ones favoured the 16 y half-life, but this matter was not persued further.

γ-ray data (Table 8) seem to be satisfactory with the possible exception of Rh-106, Ce-Pr-144 and Eu-154, where the systematic uncertainties for calculating absolute γ-ray intensities from β-branching data dominate.

For the majority of fission products only their rate of removal via neutron capture is of interest, and the important figures are cross section values and their absolute uncertainties. For fission products with very small cross sections even a 100% error may be tolerable. Therefore the cross section values (>1b) and their absolute uncertainties are displayed in Table 9.

If, on the other hand, the rate of formation of the capture product is calculated, the uncertainty of the result depends directly on the relative uncertainty of the parent's cross section. Examples are Cs-133, and fission products between Pm-147 and Eu-153 (possibly) contributing to the formation of Eu-154. In the case of radioactive nuclides within this chain (e.g. Pm-148, Pm-148m, Sm-153), where neutron capture competes with decay, both, relative and absolute cross section uncertainties are significant.

The uncertainties of even the most important cross-sections are rather high. On the other hand, the requirements depend on how well a particular neutron spectrum is known and how accurately it can be represented in calculations. Therefore sensitivity studies are recommended to define the needs.

The status of fission product yields, as presented in Table 10, is taken from the ENDF/B file [10], which is based on the compilation and evaluation of Meek and Rider [11]. Generally, the nuclides listed have cumulative yields so close to the chain yield that the latter can be used, or at least the uncertainty is the same as that of the chain yield.

One exception is the mass 135 chain, where the 2 Xenon isotopes have a relatively high independent yield. This is, however, only significant in the case of short irradiations (i.e. a few hours; e.g. in active assay techniques) or during changes in the neutron flux. Other independent yields, e.g. for Cs-134, Pr-142, Pm-148g+m and Eu-154, can be neglected as these nuclides can only be measured after their formation from neutron capture in significant amounts.

"Fast" yields in current evaluations are still a mixture of values measured in fast reactor spectra and fission neutron spectra. Generally, the yields measured in fast reactor spectra are more accurate and receive higher weight in evaluations.

4. OPEN QUESTIONS AND PROBLEMS

4.1. Calculation of accuracy requirements

Safeguards users of nuclear data should define and calculate their accuracy requirements. An appropriate way to do this are sensitivity studies, which at the same time serve as a justification for requests. I described this method in a paper presented at a recent safeguards' meeting [12]. The essential part on sensitivity studies is reproduced here as Appendix B.

4.2. Data for fast reactors

So far only needs for the safeguarding of thermal reactors have been expressed. Question: Can requirements for fast reactors be defined already at present? If yes: what are they?.

4.3 Equilibrium decay data

Most of the actinides present in nuclear fuel are produced during irradiation in a reactor. A reactor fuel lifetime is not sufficient to reach secular equilibrium with daughter products. I assume that also the naturally occuring Th-232 and Uranium isotopes are separated from (at least a part of) their daughters during the fuel fabrication process. In this event, equilibrium decay data for actinides are not needed, but a reply by safeguards experts would be useful.

4.4. Representation of neutron reaction cross sections

The replies to the questionnaire on nuclear data needs for safeguards reflected diverting opinions on the question whether one-group, few-group or energy-dependent cross-section data should be included, as illustrated in Appendix A (questions 2 and 3).

The options for spectrum averaged cross-sections are:

a) one- or few-group data
b) sets of one group data for different typical reactor spectra.

The questions to safeguards experts are:

- Are one-group data, calculated from σ_o and I (with g factors) sufficient? Possibly for a limited number of nuclei?
- Few-group data: which groups?
- Are fission spectrum averaged cross section data relevant?
- Which are "typical" reactor spectra? By which parameters can they be described?
- Who could be in a position to calculate the sets of one-group data?

For presenting $\sigma(E)$ data the following options exist:

- point cross section data
- multigroup cross section data
- resonance parameters.

Questions (see also Appendix A, question 2):

- Are $\sigma(E)$ data really needed?
- Which representation is most desirable?
- Is a special file for safeguards desirable or existing files sufficient, from which data can be retrieved upon request?

We do not consider to tabulate $\sigma(E)$ data in the handbook, since such data can only be used in large computer codes anyway. We could, however, reproduce plots of selected cross section curves for illustration. Question:

- Would this be desirable?

- If yes, which cross sections?

4.5. Spectra of prompt fission neutrons

Spectra are hard to present in numerical form. Any choice depends on the way these data are used (e.g. in calculations). Graphs serve only for illustration. Questions:

- How is the spectrum information used?

- In which form should it be presented?

$\bar{\nu}_p$ and M(n) are requested as a function of neutron energy. The most convenient way of presentation would be formulae describing the energy dependence. However, I have received no information from data experts on the existence of such formulae.

4.6. Delayed neutron data

Here the problems are: the numerical presentation of spectra and a way of describing the time dependence of the data. These questions were presented at a recent meeting on delayed neutrons [13], but no immediate solution could be proposed. We were asked to contact the experts individually with the problem.

Again, the question to safeguards experts is, how these multi-dimensional data (energy-time dependence) are used (see also Appendix A, question 4).

4.7. Prompt and delayed γ-ray data.

The problems and questions are similar to those for prompt and delayed neutrons.

4.8. γ-ray attenuation coefficients

No recommendation on available data was received yet. However, calculated values for pure elements exist and will be included since there was strong support in the replies to the questionnaire (Appendix A, question 5).

Three problems still remain:

- existence and availability of experimental data;

- definition of typical composite absorbers, for which attenuation coefficients should be measured;

- stimulation of such measurements.

Here the assistance of all interested users and experts is needed to find solutions.

4.9. Other non-neutron and macroscopic data

Information on these items does exist and can hopefully be made available to NDS (e.g. neutron source properties).

However, some more information on the needs has to be given (see section 2.4):

- availability: items b,c,d,e,h,
- presentation: all items except i,j,m,n,
- further specifications (nuclides, source, meaning, what is typical?): items: b,c,f,g,h,j (etc.?), k,l.

5. CONCLUSION

The first issue of the handbook on nuclear data for safeguards will include those data which are probably most important for safeguards users. More data, which are or will be available, will be added in 1987. However, to achieve the goal of a complete handbook that satisfies almost all safeguards users' needs, further cooperation of experts in the field of safeguards, data measurement, evaluation and application is necessary. Therefore I ask the reader to respond to the questions presented in this paper, or pass them on to other experts, or name such experts to me.

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[13] Specialists' Meeting on Delayed Neutron Properties, Birmingham, UK, 15-19 September 1986

APPENDIX A:

Extract of a questionnaire sent to safeguards experts and summary of their replies

The questionnaire started with an explanation of the handbook and its contents. The following is a literal extract of the evaluation of replies, starting with the reply to the first question on the agreement to the scope of the handbook:

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Replies: The proposal was unanimously agreed.

Comments: The handbook should not be too bulky; well indexed, so that information is easy to find.
A computer base should be designed to facilitate updating of the handbook and enable retrievals.
References to original publications should be included.

We intend to include in the handbook, for each safeguards-relevant nuclide, half lives, tables of x- and γ-ray energies and intensities and typical cross section data for thermal reactors, i.e.: 2200m/s cross-sections, Maxwellian spectrum averaged cross-sections, infinite dilute resonance integrals, Cf-252 fission spectrum averaged cross-sections (only for specific nuclei, as indicated in table 2*) by "fast"), and, if feasable and sufficient, sets of single cross-section values typical for certain reactor types.

There are also requests for point cross section data, σ(E), e.g. neutron cross-sections from 0.025 eV to 20 MeV. These data would only be available from the computer file.

Question 2: Do you in principle agree to the above suggestions of data to be included in the handbook and the data file?

Replies: The suggested data to be included were generally agreed. Additional requests were:
- decay branching ratios
- intrinsic X-ray line widths

In one reply (Harwell) doubt was expressed about the need for energy-dependent point cross section data σ(E). Specialists would be needed to develop and run the associated codes and more nuclides than indicated in Table 2-A would have to be included.

Comments: Fundamental constants only (such as σ_0, I_0,..) should be given in the first part, separated from reactor dependent spectrum averaged cross sections.

Question 3: Are sets of single value cross-sections typical for certain reactor types sufficient for safeguards purposes? Please, specify relevant cases.

Replies: Here the replies diverted, ranging from
- a rather large set of single-value spectrum averaged cross sections, even for a given reactor type, for different core locations and conditions which have to be well specified, to

*) In this paper: Table 1 on page 3

- few-group constants together with parameters characterizing the reactor spectrum, or libraries with different sets of group cross sections for different reactor types.

Comment by M. Lammer: It seems that for simple calculations typical one-group spectrum averaged cross sections are sufficient. But for more sophisticated inventory calculations, for each reactor type factors like fuel geometry, control rods, void fractions etc. have to be taken into account. For a solution to the problem, which cross section sets to include in the handbook, the assistance of experts in using cross-section codes has to be sought.

Regarding the prompt fission neutron and γ-ray data, typical values for fission in a Maxwellian or thermal reactor spectrum, etc., will be given together with formulae for the dependence on incident neutron energy, where possible.

Question 4: Do you have any suggestions on the presentation of time dependent delayed neutron and γ-ray spectra?

Replies did not bring a solution. Suggestions were:
- retain delayed neutron groups (no suggestions for delayed gammas);
- tabulations of spectra;
- plots of spectra;
- representative plots together with
- references for time-dependence, or
- approximation formulae.

Comment by M. Lammer: Plots are probably useful for inspectors but cannot be used as input to calculations. Do formulae or references for time-dependence of spectra exist? The problem is not yet solved.

The second part of the handbook will contain non-nuclear and macroscopic data, the presentation of which has still to be clarified.

Experimental data on energy-dependent γ-ray attenuation coefficients for well specified fuel types could be included as soon as they are available. Presently there exist only tabulations of calculated attenuation coefficients for single elements as absorbers.

Question 5: Should such tables of calculated values be included in the handbook or are references to pertinent publications sufficient, until experimental data described above become available?

Replies: The majority of replies supported the inclusion of calculated γ-ray attenuation coefficients for elements.

Comments: Only few replies suggested that these calculated values are sufficient; others expressed concern that experimental results for attenuation coefficients depend on fuel type and geometry and would not be representative enough for universal use. The majority of replies considered existing tabulations not sufficient, but support their inclusion as a first step. Attenuation coefficients for compounds in nuclear fuel material (experimental or calculated) would be necessary.

Question 6: Would it be valuable to present an additional table of all γ-rays included in the first part, but sorted by increasing γ-ray energy?

Replies: Again, the majority of replies supported the inclusion of such a table.

Comments: It was felt that existing γ-ray catalogues of this kind contain too many γ-lines not occuring in fresh or spent fuel.
It was further suggested to include also energies of single and double escape peaks, and data for associated γ-rays and daughter products.

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After the above summary of questions and replies follow discussions and tabulations of data requirements as presented in chapter 2 of this paper. In addition, the questionnaire contained the following survey of requirements sorted by safeguards method:

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Description of the attached table

The attached table contains suggestions of what should be included in the handbook, updated after receipt of replies. The nuclear data needed are associated with certain safeguards methods identified by numbers as follows (revised according to the replies received):

1) Active and passive neutron assay techniques
2) Gamma-ray spectrometry of fresh fuel
3) Fission product gamma-ray spectrometry of spent fuel
4) Gamma ray transmission measurements
5) X-ray fluorescence measurements
6) k-edge densitometry
7) Foil activation techniques
8) Isotopic correlations by mass spectrometry
9) Calorimetry

Table A1: Types of nuclear data used in safeguards and purpose

method	data types	purpose
1	Th-, U- and Pu- isotopes (also some isotopes of Am, Cm, Cf for simulating Pu in experiments): total, alpha and spontaneous fission half-lives, induced fission cross-sections, ν_p α-energies and intensities from decay	for evaluation of measurements and comparison with calculations of the origin of neutrons (from spontaneous fission and (α,n) reactions)
	(α,n) cross-sections for C, O, F, Al and minor elements occurring in fresh and recycled fuel	
	macroscopic data: neutron yields from spontaneous fission and (α,n) reaction per gram of isotopes concerned and second; thick target (α,n) yields per α for relevant elements and isotopes vs. α-energy and for α-spectra from decay of U-, Pu-, etc. isotopes.	
	prompt fission-neutron energy and multiplicity distributions, for neutron induced fission as well as for spontaneously fissioning nuclides	detector and counting equipment design and calibration (energy dependence, coincidence optimisation)
	prompt fission gamma-ray multiplicity distributions, yields and correlations, as above including spontaneously fissioning nuclides	as above for neutron-gamma coincidence counting
	delayed neutron and gamma-ray yields and spectra vs. time	as above for delayed coincidence counting; at short decay times also for dn and dγ spectrum measurements after activation (active interrogation)
	cross-sections for (n,x) reactions on H, D, He-3,4, Li-6, Be-9, B-10, Cd-113	neutron counters
	photoneutron production data	interfering neutrons; photoneutron sources

Table A1 (cont'd)

method	data types	purpose
	data for isotopic neutron sources (yield/g, yield/Ci, energy spectra, average energy etc.)	neutron sources for active interrogation
	library of yields, half-lives, γ-ray energies and intensities of fission products	investigation of activation buildup in fuel standards used for calibration (active assay)
	Pa, Np, Am and Cm isotopes present in spent fuel: neutron capture cross-sections for formation of these isotopes	calculation of the build-up of higher actinides in spent fuel, for comparison with measurements
2+4	gamma-ray and X-ray energies*) and intensities, half-lives for Th-, U-isotopes, Pu-isotopes, Am-241;	spectrum analysis, corrections for interference of γ-rays from minor isotopes
	the same for daughter products of these nuclides	calculation of time dependence of energy spectra in case equilibrium is not reached
3	thermal and some fast fission yields, half-lives, γ-ray energies*) and intensities, neutron capture cross-sections averaged over reactor neutron spectra, for selected fission products	spent fuel γ-spectrum analysis, interpretation of measured fission product activities and their ratios, determination of burnup
	accurate intensity values for less abundant high energy gamma-rays of fission products	to increase reliability of intrinsic calibration of spectrometers
2,3,4	total attenuation coefficients as a function of photon energy for Th-, U- and Pu- containing fuel	transmission measurements; correction for attenuation

*) Precise energy values and intrinsic line widths of X-rays are important for the analysis of composite peaks

Table A1 (cont'd)

method	data types	purpose
5+6	X-ray energies *) and intensities for Th, U, Pu; excitation cross sections, fluorescence yields	spectrum analysis
7	foil activation data, e.g.: In-115(n,n')In-115m; Ni-58(n,p)Co-58; Fe-54(n,p)Mn-54; Al-27(n,p)Mg-27; Mg-24(n,p)Na-24; Fe-56(n,p)Mn-56; Al-27(n,α)Na-24	foil activation technique
8	fission and capture cross-sections of U-233 to 236,238, Pu-238 to 242; cumulative yields from U-233,235, Pu-239,241 thermal fission and U-238 fast fission, and capture cross sections for stable isotopes of fission product Kr, Zr, Mo, Ru, Xe, Nd, Sm. half-lives of Xe-133,135.	help to resolve discrepancies between measured and calculated correlations; determination of burn-up
9	data for specific heat production	analysis of measurement results

*) Precise energy values and intrinsic line widths of X-rays are important for the analysis of composite peaks

APPENDIX B:

Sensitivity studies

Apart from meetings, the most suitable means to communicate requirements for new or more accurate nuclear data to measurers and evaluators is WRENDA, the World Request List of Nuclear Data, published every four years by NDS. However, requests to be included in WRENDA are usually screened by national authorities for their justification before they reach NDS. On the other hand, WRENDA is used by experimenters for planning and justifying their measurement programmes.

Justifications of requests can be backed up by sensitivity studies, which are quantitative calculations of the incfluence of nuclear data uncertainties on the uncertainty of the quantity to be determined.

The partial error $\Delta_i R$ of the result (of a measurement or calculation), $R = R(x_1, x_2, \ldots, x_n)$, can be given as

$$\Delta_i R = \frac{\partial R}{\partial x_i} \Delta x_i \qquad (1)$$

where the parameters $x_i \pm \Delta x_i$ are the nuclear data in our case and are assumed to be uncorrelated.

The <u>sensitivity</u> S_i of R with respect to x_i is defined by

$$\frac{\Delta_i R}{R} = S_i \frac{\Delta x_i}{x_i} \qquad (2)$$

From (1) and (2) we get

$$S_i = \frac{\partial R}{\partial x_i} \cdot \frac{x_i}{R} \qquad (3)$$

The total relative error of R contributed by nuclear data uncertainties can be calculated from

$$\frac{\Delta R}{R} = \sqrt{\sum_{i=1}^{n} \left(S_i \frac{\Delta x_i}{x_i} \right)^2} \qquad (4)$$

The analytical solution of these equations is simple and S_i can be calculated from (3) if R consists only of sums and differences ($S_i = x_i/R$) or of products and quotients ($S_i = 1$) of the x_i, but becomes tedious if R contains complex exponential functions. The best way to overcome this difficulty is to replace in computer calculations of R the input values x_i of the nuclear data by $x_i \pm \Delta x_i$ to obtain $R \pm \Delta_i R$, and calculate S_i from (2).