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Assessment and Evaluation of Decay Data for Nuclear Reactor Applications

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Discussions were initiated in 1994/95 to identify specific inadequacies in the decay data of the Joint Evaluated File (JEF) prepared by the Nuclear Energy Agency (NEA) Data Bank for nuclear reactor applications. Various correspondence has taken place within Europe between the staff responsible for this file. The net result has been the generation of lists of radionuclides that were believed to require either further detailed assessments (to check for completeness and consistency) or comprehensive re-evaluations of their decay data. These radionuclides are of direct relevance and application to either fission reactors and fuel reprocessing, or fusion research:

(a) A list of 37 fission-product radionuclides in priority order was formulated for which the decay data were judged to be of primary importance. Decay-scheme data for a further 35 radionuclides were also defined as being inadequate, whilst being particularly important in decay heat calculations.

(b) The decay data of approximately 50 radionuclides within the European Activation File (EAF) were identified as being problematic or incomplete when used for fusion reactor applications. These data exhibit inconsistencies between the mean gamma energies and component radiations (gamma rays, x-rays, annihilation radiation and bremsstrahlung).

Using a combination of subjective judgement and statistical analysis, a series of clear directives were formulated, so that further efforts could be focused on improving and

evaluating the decay data of those radionuclides judged to be inadequate. These re-evaluations are underway for completion at the end of 1998.

NUCLEAR DECAY DATA, EVALUATIONS, DECAY-DATA FILES.

1. INTRODUCTION

Decay-data files have been assembled over many years for direct application in the nuclear power industry, fuel reprocessing and waste management (Dunford, 1992; Nordborg *et al.*, 1992; Nordborg and Salvatores, 1994; Roussin *et al.*, 1994; Zongdi *et al.*, 1994). Some of these data files have been judged to be inadequate under certain circumstances, and requests have been made to re-evaluate the decay data for a number of specific radionuclides if improvements can be made (Mills *et al.*, 1995; Storrer and Nichols, 1995).

Assessments and discussions have taken place under the auspices of the Nuclear Energy Agency (NEA) Data Bank to improve the quality and consistency of the decay data in the Joint Evaluated File (JEF) assembled in Europe (Nordborg and Salvatores, 1994). Efforts within the UK have focused on the identification of those radionuclidic decay data judged as inadequate for decay-heat calculations, fuel recycling, reprocessing and fission-product standards associated with thermal fission, and activation studies identified with fusion technology. These studies are being made as part of a modest evaluation programme to assist in the development of the decay-scheme data for the Joint Evaluated Fission and Fusion file (JEFF-3) that will replace JEF.

2. THERMAL FISSION RADIONUCLIDES

A set of 37 thermal fission products were identified as important from the point of view of radiotoxicity, fuel reprocessing, monitoring standards and delayed neutron emissions (Table 1). The JEF decay-scheme data for these radionuclides have been assessed in detail to determine whether extensive re-evaluations are merited (Nordborg and Salvatores, 1994). Subjective judgements have been made, and are listed in column 4 of Table 1:

(a) decay data for ten of these radionuclides were judged to be adequately quantified to the desired detail and consistency;

(b) twenty-four radionuclides merit comprehensive re-evaluations of their decay data;

(c) three radionuclides require minor modifications to their decay data (denoted as (Yes?)).

On the basis of this preliminary review of the available data, a series of re-evaluations have begun, as listed in Table 2.

Other specific fission-product radionuclides are not included in the decay-data files of JEF, but contribute significantly to the decay heat of irradiated fuel (>0.01 of the fractional cumulative yield). These nuclides are listed in Table 3. Decay data for 33 of these radionuclides can be found in the US ENDF/B-VI file (Dunford, 1992; Roussin *et al.*, 1994), and these data have been assessed to determine their suitability for adoption in the JEFF file. All of the radionuclides have short half-lives (less than or equal to 3 sec), with Q-values and branching fractions that have been estimated for the negatron (β^-) and negatron-delayed neutron (β^-n) decay modes (apart from Nd-157 which is defined as undergoing β^- decay only). There are no decay data to be found in the ENDF/B-VI file for Sb-141 and Ce-158. Furthermore, the data for the

other radionuclides are limited to calculated average energies for the gamma-ray and beta-particle components; these data have been used to generate continuum spectra. Delayed neutron spectra were added independently from a combination of sources.

There are no obvious improvements that can be made to the ENDF/B-VI decay data unless discrete spectra can be measured for the radionuclides of concern. Rapid irradiation and chemical separation techniques are required to achieve such an objective, and a dedicated experimental programme of work would have to be established to generate discrete spectra and half-life data. Under such circumstances, all theoretical and experimental studies of these individual radionuclides need to be assessed in order to determine when their decay parameters are sufficiently well characterised for a more comprehensive evaluation to be carried out for the JEFF data file.

3. FUSION-BASED RADIONUCLIDES

The European Activation File (EAF) contains a number of radionuclides with problematic decay data (Forrest and Sublet, 1997; Table 4). These specific decay-scheme data exhibit inconsistencies and are incomplete. While approximately 50 radionuclides have been identified in this way, these evaluation requirements have been extended by including associated ground or metastable states. Decay data for forty of the activation product radionuclides have been evaluated, and the aim is to complete this exercise by the end of 1998.

4. CONSISTENCY OF DECAY DATA

The evaluated decay data included half-lives, decay energies and emission probabilities, Q-values and branching fraction(s). Details of the adopted evaluation procedures are described by Nichols (1996). Calculated decay energies were derived and compared with the sum of the decay components (e.g. beta and gamma).

$$\text{effective Q - value} = \sum_{i=1}^{\text{all BF}} Q_i BF_i$$

where Q_i and BF_i are the Q-value and Branching Fraction of the i-th decay mode;

$$\text{calculated Q - value} = \sum_{i=1}^{\text{all }\gamma} E_{\gamma_i} P_{\gamma_i} + \sum_{j=1}^{\text{all }\beta} E_{\beta_j} P_{\beta_j} + \ldots.$$

where E_{γ_i}, E_{β_j} etc and P_{γ_i}, P_{β_j} etc are the energies and emission probabilities of the i-th gamma transition, j-th beta particle transition etc of the individual decays. These values have been compared to derive the percentage deviation, which represents a quantification of the consistency of the recommended decay data (see Tables 2 and 4 for the decay data processed to date).

Consistency (defined as percentage deviation)

$$= \left[\frac{\text{(effective Q - value) - (calculated Q - value)}}{\text{(effective Q - value)}} \right] \times 100$$

Percentage deviations above 5% are regarded as high, and imply a poorly defined decay scheme; a value of less than 5% indicates the construction of a reasonably consistent decay scheme. This exercise has assisted greatly in the generation of reliable and consistent decay-scheme data, as well as eliminating various difficulties in the earlier data file.

5. CONCLUSIONS

Decay data are being derived for well-defined sets of radionuclides that are judged to be important for thermal fission and fusion applications. Efforts in 1996-98 are focused on the evaluation of decay data for 39 fission products and 74 activation products, which represent a subset of the data to be incorporated into a provisional file under preparation for the NEA Data Bank (JEFF-3).

The validity and overall consistency of the proposed decay schemes are being quantified in terms of their completeness: such tests are essential in developing credible data files on an international basis for a range of applications in the nuclear industry. Any significant difficulties in these most recent evaluations are highlighted, including any need to undertake clarifying measurements.

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Table 1: Fission Product Nuclides Requiring Well-defined Decay Data

Nuclide	Priority	Importance	Re-evaluate?
45-Rh-106	high	Instrumentation for recycling	Yes
44-Ru-106	high	Instrumentation for recycling	No
53-I-129	high	Radiotoxicity	No
54-Xe-135	high	Instrumentation for recycling	No
56-Ba-140	high	Fission product standard	No
57-La-140	high	Fission product standard	Yes
62-Sm-147	high	Instrumentation for recycling	Yes
34-Se-79	high	Radiotoxicity	Yes
40-Zr-93	high	Radiotoxicity	Yes
50-Sn-126	high	Radiotoxicity	Yes
50-Sn-117m	medium	Reprocessing	No
51-Sb-127	medium	Reprocessing	(Yes?)
52-Te-127	medium	Reprocessing	(No)
52-Te-127m	medium	Reprocessing	(No)
53-I-131	medium	Reprocessing	No
53-I-132	medium	Reprocessing	Yes
52-Te-132	medium	Reprocessing	(Yes?)
53-I-138	medium	Reprocessing/Delayed neutrons	Yes
58-Ce-141	medium	Reprocessing	No
58-Ce-144	medium	Reprocessing	No
59-Pr-143	medium	Reprocessing	Yes
59-Pr-144	medium	Reprocessing	Yes
65-Tb-161	medium	Reprocessing	Yes
35-Br-88	medium	Delayed neutron emissions	Yes
35-Br-89	medium	Delayed neutron emissions	Yes
35-Br-90	medium	Delayed neutron emissions	Yes
37-Rb-94	medium	Delayed neutron emissions	Yes
39-Y-98m	medium	Delayed neutron emissions	Yes
53-I-137	medium	Delayed neutron emissions	Yes
39-Y-99	low	Delayed neutron emissions	Yes
51-Sb-135	low	Delayed neutron emissions	Yes
53-I-139	low	Delayed neutron emissions	Yes
35-Br-87	low	Delayed neutron emissions	Yes
35-Br-91	low	Delayed neutron emissions	(Yes?)
37-Rb-95	low	Delayed neutron emissions	Yes
37-Rb-93	low	Delayed neutron emissions	Yes
33-As-85	low	Delayed neutron emissions	Yes

Table 2: Evaluated Radionuclides - Thermal Fission

Nuclide	Half-life	Consistency (% Deviation)
33-As-85	2 sec	
34-Se-79	6.0(5) x 10^5 y	0.0000
(34-Se-79m)	3.90(2) min	-0.0962
35-Br-87	56 sec	
35-Br-88	16 sec	
35-Br-89	4.5 sec	
35-Br-90	1.9 sec	
35-Br-91	0.6 sec	
37-Rb-93	5.8 sec	
37-Rb-94	2.7 sec	
37-Rb-95	0.4 sec	
(39-Y-98)	0.6 sec	
39-Y-98m	2 sec	
39-Y-99	1.5 sec	
(39-Y-99m(?))	(?)	
40-Zr-93	1.53(10) x 10^6 y	1.2384
(41-Nb-93m)	5890(50) d	-0.3678
45-Rh-106	30.1(1) sec	-0.0243
(45-Rh-106m)	132(3) min	-0.0487
50-Sn-126	2.07(21) x 10^5 y	0.0293
(51-Sb-126)	12.41(5) d	-0.0653
(51-Sb-126m)	19.1(2) min	-0.1714
(51-Sb-126n)	11(2) sec	-0.3560
51-Sb-127	3.84(3) d	-0.0431
51-Sb-135	1.7 sec	
(52-Te-127)	9.35(6) h	-0.0037
(52-Te-127m)	109(2) d	-0.0908
52-Te-132	3.23(3) d	0.1077
53-I-132	2.283(8) h	-0.0832
(53-I-132m)	83(1) min	-0.3723
53-I-137	24 sec	
53-I-138	6.4 sec	
53-I-139	2.3 sec	
57-La-140	40.285(5) h	-0.0108
59-Pr-143	13.56(1) d	0.0000
59-Pr-144	17.28(2) min	0.0382
(59-Pr-144m)	6.9(7) min	-0.0860
62-Sm-147	1.06(1) x 10^{11} y	-0.0023
65-Tb-161	6.89(3) d	-0.0324

Nuclides in parenthesis have not been requested, but were included for completeness.
Nuclides without data are awaiting evaluation.

Table 3: Important Short-lived Fission Products: Partial Summary of ENDF/B-VI Decay Data

Nuclide	Quoted Half-life (sec)	Continuum Spectra - Energy Range (keV)*		
		Gamma	Beta	Neutron
39-Y-104	0.12825	0(500) - 12730	0 - 12690	0 - 5510
39-Y-105	0.14688	0(500) - 10820	0 - 10790	0 - 6840
40-Zr-105	0.49263	0(500) - 8290	0 - 8260	0 - 2260
40-Zr-106	0.90709	0(500) - 6380	0 - 6350	0 - 2570
40-Zr-107	0.24295	0(500) - 9230	0 - 9200	0 - 3950
41-Nb-109	0.31537	0(500) - 8760	0 - 8730	0 - 5300
42-Mo-109	1.4085	0(500) - 6700	0 - 6670	0 - 1200
42-Mo-111	0.46637	0(500) - 8020	0 - 7990	0 - 2210
42-Mo-112	0.97537	0(500) - 6020	0 - 5990	0 - 2720
43-Tc-113	0.65238	0(500) - 7540	0 - 7510	0 - 4080
43-Tc-114	0 20226	0(500) - 10610	0 - 10580	0 - 4790
43-Tc-115	0.27044	0(500) - 8870	0 - 8840	0 - 5910
43-Tc-116	0.11549	0(500) - 11860	0 - 11830	0 - 6650
44-Ru-115	0.87844	0(500) - 7250	0 - 7220	0 - 1400
44-Ru-116	1.7004	0(500) - 5510	0 - 5480	0 - 2150
44-Ru-117	0.34277	0(500) -8500	0 - 8470	0 - 3180
44-Ru-118	0.66235	0(500) - 6530	0 - 6500	0 - 3680
44-Ru-119	0.19495	0(500) - 9290	0 - 9260	0 - 4440
45-Rh-118	0.31565	0(500) - 9970	0 - 9940	0 - 3410
45-Rh-120	0.17246	0(500) - 10770	0 - 10730	0 - 4830
45-Rh-121	0.24956	0(500) - 8790	0 - 8760	0 - 5990
46-Pd-121	0.64367	0(500) - 7560	0 - 7530	0 - 1520
51-Sb-141	No entry in ENDF/B-VI			
57-La-152	0.28495	0(500) - 8810	0 - 8770	0 - 3980
58-Ce-153	1.4688	0(500) - 5820	0 - 5790	0 - 1620
58-Ce-154	2.0161	0(500) - 5010	0 - 4970	0 - 1640
58-Ce-158	No entry in ENDF/B-VI			
59-Pr-156	0.37926	0(500) - 8690	0 - 8660	0 - 2790
59-Pr-157	0.38001	0(500) - 8130	0 - 8100	0 - 3590
60-Nd-157	2.4833	0(500) - 5560	0 - 5520	None
60-Nd-158	2.6949	0(500) - 5000	0 - 4970	0 - 320
60-Nd-159	0.64159	0(500) - 7150	0 - 7120	0 - 1230
60-Nd-160	0.78856	0(500) - 6350	0 - 6320	0 - 1830
61-Pm-159	3.0005	0(500) - 5650	0 - 5620	0 - 410
61-Pm-160	0.72892	0(500) - 7800	0 - 7770	0 - 1130

*Expressed in terms of incremental units of 10 keV starting from zero (first incremental energy step of continuum gamma spectra is from zero to 500 keV).

Table 4: Evaluated Radionuclides - Fusion Activation Products

Nuclide	Half-life	Consistency (% Deviation)
(25-Mn-58)	65 sec	
25-Mn-58m*	2.7 sec	
31-Ga-77	13 sec	
33-As-82	14 sec(?)	
(33-As-82m)	19 sec(?)	
34-Se-79*	$6.0(5) \times 10^5$ y	0.0000
34-Se-79m	3.90(2) min	-0.0962
38-Sr-87m	2.8 h	
39-Y-96*	5.37(7) sec	-0.0151
(39-Y-96m)	9.62(15) sec	0.0079
(39-Y-96n)	(?)	N/A
41-Nb-100	1.4(1) sec	0.0733
(41-Nb-100m)	2.9(2) sec	-0.0167
43-Tc-97*	$2.6(4) \times 10^6$ y	-0.0047
43-Tc-97m	90.2(11) d	0.0621
46-Pd-109	13.4 h	
(46-Pd-109m)	4.7 min	
46-Pd-112	20.3(2) h	-0.0306
(47-Ag-107m)	44.1(4) sec	-0.0525
(47-Ag-114)	4.5 sec	
47-Ag-114m*	2 min(?)	
(47-Ag-115)	18 sec(?)	
47-Ag-115m*	20 min(?)	
48-Cd-107	6.52(2) h	-0.0289
49-In-112	14.7(7) min	0.1052
(49-In-112m)	20.7(1) min	-0.1202
56-Ba-129	2.38(11) h	-0.0730
56-Ba-129m*	2.14(5) h	0.0550
58-Ce-147	57(2) sec	0.0269
59-Pr-143	13.56(1) d	0.0000
59-Pr-144	17.28(2) min	0.0382
(59-Pr-144m)	6.9(7) min	-0.0860
59-Pr-150	6.1(4) sec	-0.6261
(61-Pm-152)	4.12(9) min	-0.3799
(61-Pm-152m)	7.5(1) min	-0.7796
61-Pm-152n*	14.4(7) min	-0.0401
(65-Tb-156)	5.17(12) d	-0.3867
65-Tb-156m*	24.4(10) h	0.5785
65-Tb-156n*	5.1(3) h	-0.0964
67-Ho-160**	25.3(7) min	-0.2337
67-Ho-160m	5.0(1) h	-0.5027+

Table 4 (cont)

Nuclide	Half-life	Consistency (% Deviation)
67-Ho-160n**	2.9(2) sec	0.2220
67-Ho-161	2.48(12) h	0.0565
(67-Ho-161m)	6.77(6) sec	0.1297
(67-Ho-170)	43 sec(?)	
67-Ho-170m	2.8 min(?)	
72-Hf-178m	4 sec	
72-Hf-178n	30.9 y	
72-Hf-180m	5.5 h	
75-Re-191*	9.7(4) min	0.0000
75-Re-192*	6.2(8) sec	0.0566
76-Os-185	93.6 d	
(76-Os-191m)	13.1(1) h	0.0520
76-Os-195*	6.5(6) min	-0.0396
77-Ir-187	10.5 h	
(77-Ir-190)	11.8 d	
(77-Ir-190m)	1.2 h	
77-Ir-190n	3.2 h	
(77-Ir-191m)	4.9 sec	
77-Ir-191n**	5.5 sec(?)	
(77-Ir-192)	74.2 d	
77-Ir-192m	1.5 min	
(77-Ir-192n)	241 y	
77-Ir-197	5.8 min(?)	
77-Ir-197m**	8.9 min(?)	
78-Pt-193*	50(9) y	2.0682
(78-Pt-193m)	4.34(3) d	-0.3390
(79-Au-192)	5.0 h	
79-Au-192m	0.029 sec(?)	
80-Hg-199m	42.6 min	
(82-Pb-201)	9.4 h	
82-Pb-201m*	61 sec	
83-Bi-208*	$3.68(4) \times 10^5$ y	0.0635
84Po-208*	2.93(4) y	-0.0380

* No gamma lines in EAF/JEF library.

** No EAF/JEF data file.

\+ Datum for decay scheme with 124 gamma-ray transitions; when further 42 gamma rays are included in the data file that are not placed in the decay scheme, Percentage Deviation is -6.4818%.

Nuclides in parenthesis have not been requested, but were included for completeness.
N/A, not applicable (judged to be insufficient evidence for existence of nuclide).
Nuclides without Consistency (% Deviation) values are awaiting evaluation.