

UK Work Undertaken After BNFL/CEA and UKAEA Reviews of JEF-2.2 Decay Data Files: 1996-2000

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Summary

A significant fraction of the UKPADD-2 decay-data library has been incorporated into version 2 of the Joint Evaluated File (JEF-2.2, OECD-NEA Data Bank). Subsequent reviews of JEF-2.2 have highlighted the need for further developments in the recommended data for fission and fusion applications. For example, UKAEA staff involved in fusion studies identified eighty nuclides for which improvements in their evaluated decay-data files would be beneficial. Furthermore, twenty-seven fission products of importance in thermal reactor studies (plus short-lived daughters and related metastable/ground states) were judged to have inadequate decay-data files in JEF-2.2, while of importance in fuel reprocessing, as monitoring standards, or because they undergo significant delayed-neutron decay. Decay data for an additional thirty-five short-lived fission products were also considered, despite the lack of measured data; theoretical data were adopted for these specific radionuclides. Sixty-one of 80 requested fusion nuclides and all of the fission products have been evaluated, and form part of the UKPADD-6 library (which contains decay data for a total of 452 radionuclides). UKPADD-6 will become available through the OECD/NEA Data Bank after undergoing formal quality assessment.

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1. INTRODUCTION

Decay-data libraries have been assembled over many years for application in the nuclear power industry, fuel reprocessing and waste management. However, specific data files within such libraries have been judged to be inadequate under certain circumstances, and requests are regularly made to extend or re-evaluate the decay data for various radionuclides if it is believed that improvements can be made.

BNFL staff have requested improved decay-data evaluations of specific fission products on the basis of OECD-NEA discussions in 1996/97 associated with JEF-2.2, and agreed collaboration in the preparation of JEFF-3 (Joint Evaluated Fission and Fusion Library, NEA Data Bank). A set of 27 fission products of importance in thermal reactor studies (plus short-lived daughters and related metastable/ground states) were identified as important from the point of view of radiotoxicity, fuel reprocessing, monitoring standards and delayed-neutron emissions (Table 1), and a lack of adequate decay-data files. The initial aim was to produce recommended decay data for the UKPADD-6 Library (1) that will subsequently be reviewed and used in the assembly of JEFF-3. Hence, a three-year evaluation exercise has been underway since mid-1996 to produce a comprehensive set of recommended decay data for these nuclides based on a well-defined evaluation procedure. Theoretical data for a further 35 short-lived fission products have also been considered for adoption from other sources (Table 2).

The main source of decay data to model specific consequences of fusion activation is JEF-2.2. The decay-data files of 50 specific radionuclides have been identified by R A Forrest (UKAEA) as being incomplete when used for fusion applications. This list of problematic radionuclides was extended to approximately 80, with the addition of related metastable states and daughter nuclides. Some of the JEF-2.2 decay-data files do not contain any gamma-ray emissions, while others exhibit inconsistencies between the mean gamma energies and component radiations listed in the files. Comprehensive decay schemes for sixty-one of these radionuclides have been evaluated, and the resulting data files incorporated into UKPADD-6.

2. EVALUATION METHODOLOGY

A logical evaluation methodology has been adopted for radionuclides with discrete and well-characterised decay parameters in order to achieve the necessary consistency and quality of the recommended data. The decay scheme is initially constructed from a combination of the various data sources. If necessary, weighted adjustments are made in order to obtain a reasonable degree of consistency, while any changes that such modifications might produce in the proposed decay scheme are noted. Thus, the decay data are cycled through a series of adjustments while maintaining satisfactory agreement with the published measurements and specific theoretical data.

Every effort is made to ensure that there is a reasonable emission-probability balance between the population and de-population of all excited levels in the decay scheme. All decay modes of each radioactive nuclide have to be completely defined in terms of the various branching ratios and the Q-values. Furthermore, the sum of all α , β^- , β^+ /electron-capture and isomeric transition probabilities are produced to be consistent with the corresponding branching ratios.

The gamma-ray emission probabilities are the photon probabilities per disintegration, and are listed as percentages in the data files. Internal conversion coefficients for a given gamma-ray transition have to be consistent with both the photon and total transition probabilities [ie. (photon + conversion electron) emission probabilities = total transition probability]. When the internal conversion of a gamma-ray transition is significant, theoretical internal conversion coefficients have been recommended if experimental data are unavailable. This process ensures that the transition energy is appropriately shared between the electromagnetic and electron components.

The transition types of all of the beta emissions are taken into account in the calculation of the mean beta energy from the evaluated end-point energies and emission probabilities; this information is inferred from the spin and parity assignments proposed for the levels involved. Energies and intensities of conversion electrons, Auger electrons and x-ray radiation are derived in a consistent manner. Finally, uncertainties are quantified for all parameters.

Each evaluation is fully documented, and the resulting file (ENDF-6 format) contains a Comments Section that includes observations of inconsistencies and describes the assumptions made by the evaluator to deal with any problems associated with that specific radionuclide.

3. THEORETICAL DECAY DATA

Thirty-five short-lived fission products are believed to contribute significantly to the early decay heat of irradiated fuel from thermal reactors (Table 2). Unfortunately, all of these radionuclides have poorly characterised decay parameters (due to a lack of measured decay data), and therefore US ENDF/B-VI decay-data files containing theoretical data for 33 of these fission products have been considered (2). These are possible candidates for incorporation into UKPADD-6. When judged to be appropriate, these files have been adopted in conjunction with various supportive data (eg half-lives, spin/parity and mean decay energies).

Part of the data-file production procedure involves using the FIZCON code (3) to assess the validity and consistency of the various recommended parameters. This process includes a check to ensure that the data set does not include any delayed neutrons with energies greater than the energy available via the decay mode ($Q(\text{beta-n})$). Although several files adopted from the US ENDF/B-VI library failed this test, resolution of this inconsistency proved to be trivial, apart from the data for two radionuclides ($^{40}\text{Zr-105}$ and $^{42}\text{Mo-109}$). The EVAP computer code was written to generate evaporation spectra based on the theory of Brady (4), and this program was applied to both problematic nuclides to produce the desired form of neutron emission spectra.

Decay data for $^{51}\text{Sb-141}$ and $^{58}\text{Ce-158}$ were calculated in a more simplified manner from the relevant $Q(\text{beta})$ values derived by extrapolation of equivalent data for adjacent nuclides. Only mean beta, gamma and alpha data and their estimated uncertainties were determined for these two radionuclides, along with half-life, spin and parity.

4. CONSISTENCY OF DECAY DATA

The evaluated decay data include half-lives, decay energies and emission probabilities, Q-values and branching fraction(s), as previously described by Nichols (5). Calculated decay energies were derived and compared with the sum of the decay components:

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

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

$$\text{calculated Q - value} = \sum_{i=1}^{\text{all } g} E_g P_g + \sum_{j=1}^{\text{all } b} E_{b_j} P_{b_j} + \dots$$

where E_g , E_{b_j} etc. and P_g , P_{b_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:

Consistency (also defined as percentage deviation)

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

Percentage deviations above 5% are regarded as high, and imply an ill-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 files. Percentage deviations are listed in Table 3 for all requested fission products for which discrete decay data have been measured (including short-lived daughters and related metastable (or ground) state radionuclides). A similar listing for 61 of the 80 requested fusion nuclides is given in Table 4. Unfortunately, this procedure could not be applied to the 35 short-lived fission products with non-discrete decay data.

5. PRODUCTION PROCEDURE

Specific difficulties were experienced in preparing the UKPADD-6 files of the delayed-neutron emitters in ENDF-B6 format. Hence, a well-defined procedure was developed in order to minimise errors between the evaluation and production of the files in ENDF-B6 format. A completely automatic approach proved

impossible to adopt because of the inability of COGEND (6, 7) to process delayed-neutron emissions; COGEND should be enhanced as soon as possible to eliminate this inadequacy.

When no experimental delayed-neutron data were available for a particular radionuclide, Brady (4) used an evaporation model to calculate theoretical delayed-neutron spectra. After a rigorous assessment, EVAP was written to calculate the delayed-neutron emission for any nuclide, given the mass (amu) and $Q(\beta\text{-n})$. The output is in ENDF-B6 format, with each energy increment (eV) paired against the probability (3 pairings per 66 field record). Material number, appropriate file (MF=8) and reaction (MT=457) are appended to each record, leaving sequence numbers to be added when the data are inserted into the evaluated file. Preservation of the overall integral to unity requires appropriate scaling from MeV to eV. Resulting spectra have been validated by application to nuclides with well-defined experimental data.

The output represents the delayed-neutron spectrum for any radionuclide, particularly those nuclides for which discrete delayed-neutron emission energies and probabilities are not known. Some of the initial data parameters in ENDF-B6 format have to be altered by hand, and sequence numbers added. Total delayed-neutron emission energy per decay is an important quantity, and two codes have been written to derive this quantity (ie., from continuous spectra (NEUTRAN) or from discrete emissions (DNMEAN)).

Both NEUTRAN and DNMEAN have been applied to specific decay-data evaluations to complete decay schemes and to help validate existing data when these parameters are taken from other sources. Such efforts to produce recommended delayed-neutron data are relatively labour intensive, and consideration needs to be given to an updating and overhaul of COGEND so that this powerful code can be improved further to process delayed-neutron decay data (ie., incorporate EVAP, NEUTRAN and DNMEAN).

6. CONCLUSIONS

A series of decay-data evaluations have been completed in a programme of work specified by staff at BNFL plc (R W Mills) and CEA (F Storrer) in preparation for the assembly of UKPADD-6 and JEFF-3. A list of 37 radionuclides evolved for discrete decay-data evaluation as a consequence of a review of the requirements of the nuclear industry with respect to decay heat, recycling, reprocessing and delayed-neutron emissions; after further assessment, these evaluation needs were reduced to 27 radionuclides (plus short-lived daughters and related metastable/ground states). The decay data for these radionuclides have been evaluated (see Table 3 for consistency of the recommended data), along with the assessment and evolution of recommended decay data for a further 35 short-lived fission products for which there are no known measurements (Table 2).

During the studies outlined above, methods were developed to assist in the evaluation of the $\beta\text{-n}$ decay mode, and delayed-neutron emissions in particular. Discrete delayed-neutron data were adopted when the appropriate measurements had been reported in the literature. Where these data were unavailable, a calculation route was used to produce continuous spectra based on the theory of Brady (4). However, the procedures adopted to generate the recommended data for delayed-neutron emitters proved to be labour intensive; this form of data-production exercise needs to be improved by developing the COGEND code (6, 7) to check and use the evaluated neutron data automatically. Further details of these most recent evaluations are given in reference 8.

A similar comprehensive evaluation exercise has been undertaken for 61 of 80 fusion nuclides requested by R A Forrest (UKAEA Fusion, Culham (Table 4)). JEF-2.2 decay-data files for these radionuclides were judged to be either inadequate or incomplete for fusion applications. The resulting recommended decay-data files for the sixty-one radionuclides have also been incorporated into UKPADD-6.

All of the evaluated data have been assembled with other decay-data files from earlier versions of UKPADD to create a new library in ENDF-6 format consisting of files for 452 radionuclides, UKPADD-6. Rigorous consistency checks have been made to confirm the validity and completeness of the data before releasing the updated library.

7. Acknowledgements

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8. References

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Table 1: Fission Product Nuclides - Requested Decay Data

Radionuclide	Priority	Application/Requirement
45-Rh-106*	high	Instrumentation for recycling
57-La-140	high	Fission product standard
62-Sm-147	high	Instrumentation for recycling
34-Se-79*	high	Radiotoxicity
40-Zr-93*	high	Radiotoxicity
50-Sn-126*	high	Radiotoxicity
51-Sb-127*	medium	Reprocessing
53-I-132*	medium	Reprocessing
52-Te-132	medium	Reprocessing
53-I-138	medium	Reprocessing/Delayed neutron emissions
59-Pr-143	medium	Reprocessing
59-Pr-144*	medium	Reprocessing
65-Tb-161	medium	Reprocessing
35-Br-88	medium	Delayed neutron emissions
35-Br-89	medium	Delayed neutron emissions
35-Br-90	medium	Delayed neutron emissions
37-Rb-94	medium	Delayed neutron emissions
39-Y-98m*	medium	Delayed neutron emissions
53-I-137	medium	Delayed neutron emissions
39-Y-99	low	Delayed neutron emissions
51-Sb-135	low	Delayed neutron emissions
53-I-139	low	Delayed neutron emissions
35-Br-87	low	Delayed neutron emissions
35-Br-91	low	Delayed neutron emissions
37-Rb-95	low	Delayed neutron emissions
37-Rb-93	low	Delayed neutron emissions
33-As-85	low	Delayed neutron emissions

*Additional short-lived daughters and related metastable/ground state radionuclides were also evaluated.

**Table 2: Short-lived Fission Products - Adoption of US ENDF/B-VI
Decay Data Unless Stated Otherwise**

Radionuclide	Quoted Half-life (sec) - USENDF/B-VI	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 ^ψ
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 US ENDF/B-VI; other theoretical data adopted			
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 US ENDF/B-VI; other theoretical data adopted			
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).

⁺Neutron spectrum adjusted to 0-1794 keV.

^ψNeutron spectrum adjusted to 0-620 keV.

Table 3: Comprehensive Evaluations - Discrete Data Sets for Fission Products

Radionuclide	Consistency (% Deviation)	Radionuclide	Consistency (% Deviation)
33-As-85	0.0988 [*]	(51-Sb-126)	-0.0653
34-Se-79	0.0000	(51-Sb-126m)	-0.1714
(34-Se-79m)	-0.0962	(51-Sb-126n)	-0.3560
35-Br-87	-0.1976 [*]	51-Sb-127	-0.0431
35-Br-88	0.2554 [*]	51-Sb-135	-0.0198 [*]
35-Br-89	0.0534 [*]	(52-Te-127)	-0.0037
35-Br-90	0.1331 [*]	(52-Te-127m)	-0.0908
35-Br-91	0.0274 [*]	52-Te-132	0.1077
37-Rb-93	-0.0182 [*]	53-I-132	-0.0832
37-Rb-94	-0.0527 [*]	(53-I-132m)	-0.3723
37-Rb-95	-0.2394 [*]	53-I-137	0.1276 [*]
(39-Y-98)	-0.0432 [*]	53-I-138	-0.1955 [*]
39-Y-98m	-0.2944 [*]	53-I-139	-0.0552 [*]
39-Y-99	-0.0741 [*]	57-La-140	-0.0108
40-Zr-93	1.2384	59-Pr-143	0.0000
(41-Nb-93m)	-0.3678	59-Pr-144	0.0382
45-Rh-106	-0.0243	(59-Pr-144m)	-0.0860
(45-Rh-106m)	-0.0487	62-Sm-147	-0.0023
50-Sn-126	0.0293	65-Tb-161	-0.0324

Additional short-lived daughter and related metastable/ground state radionuclides are in parenthesis, and were also evaluated.

^{*}Beta-decay mode only.

Table 4: Evaluated Decay Data - Fusion Activation Products (1996-2000)

Radionuclide	Half-life	Consistency (% Deviation)
7-N-17	4.17(4) sec	0.0724†
(25-Mn-58)	65.2(5) sec	-0.3450
25-Mn-58m*	2.7(6) sec	0.1037
31-Ga-77	13 sec	
33-As-82	14 sec(?)	
(33-As-82m)	19 sec(?)	
34-Se-79*	6.0(5) x 10 ⁵ y	0.0000
34-Se-79m	3.90(2) min	-0.0962
38-Sr-87m	2.808(6) h	-0.0154
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) x 10 ⁶ y	-0.0047
43-Tc-97m	90.2(11) d	0.0621
46-Pd-109	13.46(1) h	0.0090
(46-Pd-109m)	4.71(3) min	0.0367
46-Pd-112	20.3(2) h	-0.0306
(47-Ag-107m)	44.1(4) sec	-0.0525
(47-Ag-109m)	39.8(2) sec	-0.1869
(47-Ag-114)	4.7(1) sec	-0.1883
47-Ag-114m*	0.00150(5) sec	0.0039
(47-Ag-115)	20.5(4) min	0.0434
47-Ag-115m*	18.6(8) sec	0.1918
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): Evaluated Decay Data - Fusion Activation Products (1996-2000)

Radionuclide	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	4.0(3) sec	0.0013
72-Hf-178n	31(1) y	0.1450
72-Hf-180m	5.5(1) h	-0.2523
75-Re-191*	9.7(4) min	0.0000
75-Re-192*	6.2(8) sec	0.0566
76-Os-185	93.8(9) d	0.0570
(76-Os-190m)	9.9(4) min	0.0011
(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)	12.0(2) d	-0.1063
(77-Ir-190m)	1.120(3) h	-0.0849
77-Ir-190n	3.087(12) h	0.0893
(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
(78-Pt-197)	18.3 h	
(78-Pt-197m)	94.4 min	
(79-Au-192)	5.0 h	
79-Au-192m	0.029 sec(?)	
(79-Au-197m)	7.8 sec	
80-Hg-199m	42.1(9) min	0.0441
(82-Pb-201)	9.4 (1) h	0.3405
82-Pb-201m*	61(3) sec	0.1220
83-Bi-208*	3.68(4) x 10 ⁵ y	0.0635
84-Po-208*	2.93(4) y	-0.0380

† Beta-decay mode only.

* No gamma lines in EAF/JEF library.

** No EAF/JEF data file.

+ Addition of 42 gamma rays not placed in the decay scheme gives Consistency of -6.4818%.

N/A, not applicable (judged to be insufficient evidence for existence of nuclide).

Nuclides in parenthesis have not been requested, but were included for completeness.

Nuclides without Consistency values are awaiting evaluation.