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Assessment of Decay Data Files for Selected Radionuclides, Interim Report, April 1996

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1. Introduction

Discussions were initiated in mid-1995 to focus the limited amount of UK evaluation effort on improving the quality of the decay data for a specific number of radionuclides of particular interest to the nuclear industry. Lists of problematic nuclides were assembled that were initially based on discussions involving the JEF Working Group on Fission Yields and Radioactive Decay Data (1). Communications were also initiated with staff at CEN Cadarache (2) and BNFL Sellafield (3), and a series of agreed tabulations were assembled.

A list of 37 radionuclides in priority order was formulated for which the decay-data files need to be assessed in order to judge whether a detailed re-evaluation needs to be undertaken (Table 1). Decay data for a further 35 radionuclides were defined as being important in decay heat and inventory calculations, although their measured data are known to be extremely sparse (Table 2). Detailed assessments have been made of the decay data to be found in JEF-2.2 (and ENDF/B-VI data library) for all of these radionuclides(4,5). Furthermore, a decay-data checking code has been written prior to determining the overall consistency of the available files. Using such a combination of subjective judgement and statistical analysis, a series of clear directives can be produced to determine whether further labour-intensive efforts are required to improve the specific decay-data files (and hopefully generate more consistent decay schemes for these radionuclides).

2. Checking Code for Decay Data (OKDK)

A computer code named OKDK has been written to validate decay-data files in ENDF-6 format (6). OKDK can be applied to both existing and newly evaluated files, and allows the evaluator to assess the completeness of each evaluation. A full description of the current version of OKDK is included in a separate document which will be issued soon.

Average decay energies for each decay mode are calculated from the individual quanta, and compared with estimates derived in the evaluation. Two forms of total decay energy (Q-value) are considered:

(a) Effective Q-value is derived by summing the product of the Q-Value of the decay mode ($\alpha,\beta^-,EC/\beta^+,IT\ldots$) and the associated branching fraction.

(b) Calculated Q-value is determined by summing the product of the particle energy and emission probability for all emitted particles.

These two estimates of total decay energy are then compared, and the results are displayed.

OKDK can be used in conjunction with the existing decay data checking codes (CHECKR and FIZCON (7)) to form part of the quality assurance validation for the assembly of the evaluated decay data in the Joint Evaluated Fission and Fusion file (JEFF3.0).

Comprehensive definitions of the energy equations are given Section 3 below, although these definitions do not recognise the limitations of ENDF-6 format. OKDK is limited to checking:

$$\overline{E}_{"\gamma"} = \overline{ER_{\gamma}} + \overline{ER_{X-ray}},$$

noting that any discrepancy may be due to including the internal bremsstrahlung energy in the $\overline{E}_{"\gamma"}$ term since this parameter is not present elsewhere in the ENDF-6 files,

$$\overline{E}_{"\beta"} = \overline{ER_{\beta^-}} + \overline{ER_{\beta^+}} + \overline{ER_{e^-}},$$

noting the zero contribution from electron capture, and

$$\overline{E}_{"\alpha"} = \overline{ER_{\alpha}} + \overline{ER_{n}} + \overline{ER_{SF\ fragment}} + \overline{ER_{p}}.$$

The ER terms are the mean energies associated with each emitted radiation type defined as STYPE in the ENDF-6 format. It is important to note that there is a lack of neutrino/antineutrino energies in these equations.

The main function of OKDK is to check the overall Q-value or energy release (including neutrino/antineutrino energies). These ENDF-6 evaluations contain Q-values and Branching Fractions (BF) associated with each mode of decay (RTYPE) that are used to form:

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

The overall Q-value can also be calculated by adding the total energy release (including neutrino/antineutrino energies) from each emitted radiation type (STYPE):

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$$\text{Calculated Q-value} = \sum_{i=1}^{all\,\gamma} E_{\gamma_i} P_{\gamma_i} + \sum_{j=1}^{all\,\beta^-} E_{\beta_j^-} P_{\beta_j^-} + \sum_{k=1}^{all\,\beta^+\,and\,ec} E_{\beta_k^+/ec_k} P_{\beta_k^+/ec_k} \cdots$$

$$\cdots + \sum_{l=1}^{all\,e} E_{e_l} P_{e_l} + \sum_{m=1}^{all\,x-rays} E_{x_m} P_{x_m}$$

where the E terms are transition energies associated with each emitted quantity (STYPE), and the P terms are the product of the relative intensity (RI_{STYPE}) and discrete normalisation factor (FD_{STYPE}). The emitted energy for electron capture is included in both the third **and** the last two terms of the above equation. While the first term includes the neutrino energy, the final two terms constitute the electron and x-ray components that may be present as part of this decay mode.

OKDK derives the Calculated Q-value by considering each mode of decay (RTYPE):

$$\text{Calculated Q-value} \approx \sum_{RTYPE=1}^{7} Y_{RTYPE}$$

where the yield terms (Y) are defined below for each decay mode.

(a) β^- emitter, RTYPE = 1

$$Y_{\beta^-} = \sum_{i=1}^{all\,\beta^-} (E_{\beta^- + \bar{\nu}})_i P_{\beta_i^-} + \sum_{j=1}^{all\,\gamma} E_{\gamma_j} TP_{\gamma_j}$$

where $\bar{\nu}$ represents the antineutrino, and $TP_{\gamma_j} = P_{\gamma_j}[1 + ICC_{\gamma_j}(\text{total})]$ in which ICC_{γ_j}(total) is the total internal conversion coefficient for each gamma ray, defined as $RICC_{\gamma_j}$ in STYPE = 0 data of the ENDF-6 format.

(b) β^+ emitter, RTYPE = 2

$$Y_{\beta^+} = \sum_{i=1}^{all\,\beta^+} (E_{\beta^+ + \nu})_i P_{\beta_i^+} + \sum_{j=1}^{all\,\gamma} E_{\gamma_j} TP_{\gamma_j}$$

where ν represents the neutrino, and TP_{γ_j} is defined above in (a).

(c) ec decay, RTYPE = 2

$$Y_{ec} = \sum_{i=1}^{all\,ec} (E_{ec}^{max})_i P_{ec} + \sum_{j=1}^{all\,\gamma} E_{\gamma_j} TP_{\gamma_j}$$

where "max" represents the transition energy including the neutrino.

(d) α emitter, RTYPE = 4

$$Y_\alpha = \sum_{l=1}^{all\ \alpha}(E_{\alpha_i} + E_{R_i})P_{\alpha_i} + \sum_{j=1}^{all\ \gamma} E_{\gamma_j}P_{\gamma_j} + \sum_{k=1}^{all\ e} E_{e_k}P_{e_k} + \sum_{l=1}^{all\ x\,rays} E_{x_l}P_{x_l}$$

where R is the recoiling nucleus whose energy excludes that of any subsequent γ rays and electrons.

(e) RTYPE = 3, 5, 6 or 7

The isomeric transition (IT, TYPE = 3), neutron emission (n, RTYPE = 5), spontaneous fission (SF, RTYPE = 6) and proton emission (p, RTYPE = 7) decay modes have yet to be included in OKDK and validated.

The lack of a consistency check on the energies of individual discrete electrons (STYPE 8) and x-rays plus annihilation radiation energies (STYPE 9) should be noted when formulating the Q-value equations. OKDK has thus been coded to list the average energy (ER) of each radiation type (STYPE) to compare with the sum of the "discrete energy" times the product of relative intensity (RI_{STYPE}) and discrete normalisation factor (FD_{STYPE}):

$$\overline{ER_p} = \sum_{i=1}^{all\ p} E_i P_i \,.$$

There are discrepancies in these calculations for β decay and electron capture (STYPE 1 and 2) due to the lack of neutrino/antineutrino energy in the ER term.

The development of OKDK has shown that some aspects of the ENDF-6 format could be improved to assist in quality assurance checks. Both the neutrino and antineutrino energies need to be included in the format, as well as the internal bremsstrahlung energy (the latter can only be determined in the ENDF-6 format by subtracting large terms, which can lead to significant relative errors). There are also considerable variations in the decay energies, which leads to precision problems in the consistency checks and could result in the errors for small quantities going unnoticed. A further point of interest is the overall precision of COGEND(8) in generating such data. The excellent Comments Section to be found in the COGEND-generated files includes data that are quoted to more significant figures than justified in their calculation.

3. <u>Definition of Decay Parameters Within ENDF-B6 Format</u>

Much of the data contained in JEF-2.2 and ENDF/B-VI, including individual gamma-ray, beta-particle, alpha-particle, discrete electrons and x-rays, require no further explanation. However, specific features of the library have caused some confusion and are defined below, i.e., the average energy data and their components.

(a) Average Beta Energy (Average Light Particle Energy)

The average beta or light particle energy per decay is defined as the average energy of all electron emissions such as β^-, β^+, conversion electrons and Auger electrons:

$$\bar{E}_{"\beta"} = \sum_{i}^{\text{all }\beta^-} \bar{E}_{\beta_i^-} I_{\beta_i^-} + \sum_{j}^{\text{all }\beta^+} \bar{E}_{\beta_j^+} I_{\beta_j^+} + \sum_{k}^{\text{all Auger}} \bar{E}_{A_k} I_{A_k} + \sum_{l}^{\text{conv}} \bar{E}_{c_l} I_{c_l}$$

where $\bar{E}_{\beta_i^-}$, $\bar{E}_{\beta_j^+}$, $\bar{E}_{A_k}$ and $\bar{E}_{c_l}$ are the mean negatron, positron, Auger electron and conversion electron energies of the i, j, k and 1-th transition of each type respectively, and $I_{\beta_i^-}$, $I_{\beta_j^+}$, I_{A_k} and I_{c_l} are the corresponding absolute fractional intensities per disintegration.

(b) Average Gamma Energy (Average Electromagnetic Energy)

All electromagnetic radiations such as gamma rays, x-rays, annihilation radiation, and internal bremsstrahlung are included within the average gamma energy:

$$\bar{E}_{"\gamma"} = \sum_{i}^{\text{all }\gamma} \bar{E}_{\gamma_i} I_{\gamma_i} + \sum_{j}^{\text{all x-rays}} \bar{E}_{x_j} I_{x_j} + \sum_{k}^{\text{all }\beta^+} 1.022\ I_{\beta_k^+} + \sum_{l}^{\text{all }\beta^+\beta^-} \bar{E}_{\beta_l} I_{\beta_l}$$

where $\bar{E}_{\gamma_i}$ and $\bar{E}_{x_j}$ are the mean gamma and x-ray energies of the i and j-th transition of each type respectively, I_{γ_i} and I_{x_j} are the corresponding fractional intensities per disintegration (I_{γ_i} is the unconverted (observed) photon intensity), $\bar{E}_{\beta_l}$ is the mean internal bremsstrahlung energy of beta transition 1 which has an absolute fractional intensity I_{β_l} , and $I_{\beta_k^+}$ is the absolute fractional intensity of positron transition k. Note that the energy identified with the internal bremsstrahlung is included within this parameter, but does not appear elsewhere in the ENDF-6 format.

(c) Average Alpha Energy (Average Heavy Particle Energy)

The average heavy particle energy includes the mean energy of alpha particles, recoil nuclei, protons, neutrons and spontaneous fission fragments.

$$\bar{E}_{"\alpha"} = \sum_{i}^{\text{all }\alpha} \bar{E}_{\alpha_i} I_{\alpha_i} + \sum_{j}^{\text{all recoil}} \bar{E}_{R_j} I_{R_j} + \sum_{k}^{\text{all protons}} \bar{E}_{p_k} I_{p_k} + \sum_{l}^{\text{all neutrons}} \bar{E}_{n_l} I_{n_l} + \sum_{m}^{\text{fiss frag}} \bar{E}_{F_m} I_{F_m}$$

where $\overline{E}\alpha_i, \overline{E}_{R_j}, \overline{E}_{p_k}, \overline{E}_{n_l}$ and $\overline{E}_{F_m}$ are the mean alpha, recoil nucleus, proton, neutron and fission fragment energies of the i, j, k, l and m-th component of each type respectively, and $I_{\alpha_i}, I_{R_j}, I_{p_k}, I_{n_l}$ and I_{F_m} are the corresponding absolute fractional intensities per disintegration.

4. Assessment of Decay Data: Comments

Studies were made of the decay data for each individual radionuclide in Table 1 prior to agreeing on any re-evaluation exercise. The primary aim was to identify those specific radionuclides requiring further study so that the JET-2.2 datafiles can be improved in 1996/97(4).

Efforts were also made to assess any available files quantifying the decay schemes of the radionuclides listed in Table 2. Such an exercise proved to more difficult to achieve, emphasising the need for the preparation of these radioactive sources and specialised measurements of their decay parameters.

4.1 45-Rh-106

Rh-106 undergoes β^- decay with a half-life of 29.80(8) sec to the nuclear levels of Pd-106. It should also be noted that there is a metastable state of Rh-106 (half-life of 131(2) min) which also decays by beta-particle emission to Pd-106. The ground state of Rh-106 is believed to decay via 30 beta-particle emissions and 88 gamma-ray transitions, while Rh-106m decays via eight beta-particle emissions and 41 gamma-ray transitions.

The JEF-2.2 decay-data file for Rh-106 contains 28 beta-particle emissions (all assigned to Rh-106) and 95 gamma rays (some that can be assigned to Rh-106m rather than Rh-106). All of the average energy terms are in agreement and consistent. Only a limited number of the gamma-ray transitions have been assigned internal conversion coefficients, although some of these are the most important emissions in this decay mode.

A re-evaluation of the decay-data for Rh-106 would appear to be necessary in order to produce a fully consistent dataset. Furthermore, efforts should also be made to evaluate and recommend an equivalent set of comprehensive decay data for Rh-106m, and so achieve the desired consistency for both radionuclides.

4.2 44-Ru-106

Ru-106 undergoes β^- decay directly to the ground state of Rh-106, with a Q(beta)-value of only 39.4(2)keV. The JEF-2.2 file for this radionuclide reflects the simple decay scheme, and there is no requirement to re-evaluate the measured data.

4.3 53-I-129

I-129 undergoes β^- decay directly to the first excited state of Xe-129 (39.58keV nuclear level energy) prior to gamma-ray emission to the ground state. Both the beta particle and the gamma-ray transition have been well-defined, and all of the relevant data are included in the JEF-2.2 file. A detailed re-evaluation does not appear to be necessary at the present time.

4.4 54-Xe-135

Xe-135 undergoes relatively simple β^- decay to Cs-135 nuclear levels (particularly 249.77 keV level), with the emission of five beta particles and 13 gamma rays. The most significant internal conversion coefficients are included in the JEF-2.2 file for this radionuclide. All of the decay data within the file are consistent, and therefore a re-evaluation does not appear to be necessary at present.

4.5 56-Ba-140

Ba-140 has been shown to decay via 5 beta-particle and 13 gamma-ray transitions to La-140. These data are well-defined in JEF-2.2, including the internal conversion coefficients of all of the gamma rays. All of the data are consistent, and there is no obvious requirement to undertake a re-evaluation of the decay scheme for Ba-140.

4.6 57-La-140

La-140 undergoes β^-decay to Ce-140, with the emissions of 18 beta particles and 39 gamma-ray emissions. Although the majority of these data are included within the JEF-2.2 file, there are some omissions that could be addressed by a comprehensive re-evaluation. While these modifications are judged to be marginal, such an exercise would improve the database with the addition of the missing beta particles, gamma rays and internal conversion coefficients.

4.7 62-Sm-147

Sm-147 undergoes alpha decay directly to the ground state of Nd-143, with a half-life of 10^{11} years. A Q(alpha)-value of 2310.6(11)keV results in an alpha-particle energy of 2247.7kev, which does not correspond to the value of 2233(5)keV to be found in the JEF-2.2 file. Minor adjustments in these data would be beneficial, and therefore re-evaluation is recommended.

4.8 34-Se-79

Se-79 undergoes β^- decay (Q(beta)-value 150.7 keV), with an ill-defined half-life of $\leq$ 6.5×10^4 years that is based on studies reported in an Oak Ridge National Laboratory report published in 1949. Decay is rightly assumed in JEF-2.2 to go directly to the ground state of Br-79, and therefore there is virtually no gamma component (no gamma rays, x-rays, internal bremsstrahlung or annihilation radiation). Therefore, it is somewhat surprising to find that when the data were compiled at the NEA Data Bank (July 93), an average gamma energy of 33% of the Q(beta)-value was inserted into the file. This assignment is incorrect, and was made in an arbitrary manner because the original average energy terms took no account of the neutrino energy that accompanies beta decay.

The recommended data are approximately 13 years old, and a re-evaluation should be undertaken that does not result in the artificial creation of a nonsensical value for the mean gamma energy.

4.9 40-Zr-93

Zr-93 undergoes β^- decay (Q(beta)-value of 90.5 keV) to both the ground and metastable states of Nb-93 (stable and 15.8 year half-life, respectively). Although a relatively simple decay scheme (with two beta-particle emissions assigned to Zr-93, and the resulting gamma ray assigned to Nb-93m), there are some discrepancies in the JEF-2.2 datafile: Q(beta)-value of 62 keV is to the Nb-93m state only (ignoring any direct decay to the ground state), only one beta-particle emission (ignoring decay to the ground state), and the arbitrary assignment of a mean gamma energy of 20.7 keV (i.e. based on 33% of the Q(beta)-value). All of these assignments are questionable, and a re-evaluation of the relatively simple decay data is recommended.

4.10 50-Sn-126

Sn-126 undergoes β^- decay to the 11 sec and 19 min metastable states of Sb-126, and both decay modes are accompanied by gamma-ray emissions (21.65, 23.28, 42.64, 64.28, 86.94 and 87.57keV transitions). These data and the necessary internal conversion coefficients for the gamma rays are included in the JEF-2.2 file.

There are some minor inconsistencies in the recommended Q(beta)-values, the single beta-particle emission and the gamma-ray transitions. A re-evaluation of the decay data for Sn-126 is recommended, along with the daughter radionuclides (Sb-126m and Sb-126n).

4.11 50-Sn-117m

Sn-117m undergoes isomeric transition decay to the ground state (Q(IT)-value of 314.58(4)keV and half-life of 13.60(4) days), with the emission of three gamma rays. The data in the JEF-2.2 file are consistent with the simple decay scheme (apart from the 314.3keV gamma-ray transition which has a very low emission probability of 4.9 x 10^{-6}, effectively negligible). Both the average beta (light particle) and gamma (electromagnetic) energies are in good agreement with the relevant average decay energies for the radiation types. A re-evaluation of the decay data for Sn-117m is not required.

4.12 51-Sb-127

Sb-127 decays to the ground and metastable states of Te-127, with the emission of 15 beta particles and 36 gamma rays. All of these data and consistent Q-values are intended in the JEF-2.2 file, although the decay-mode definitions of 14 of the gamma-ray transitions have not been specified (i.e. via β^- decay). The average beta (light particle), gamma (electromagnetic) and alpha (heavy particle (zero)) energies are also in agreement with the average decay-type energies.

A comprehensive re-evaluation of Sb-127 does not appear to be necessary, apart from defining the originating decay mode for all of the gamma rays as beta-based.

4.13 52-Te-127

Te-127 decays with a half-life of 9.35(7)h to the ground state of I-127. Five beta particles and nine gamma-ray transitions are involved that are identifiable with a Q(beta)-value of 694(5)keV. These data are included in the JEF-2.2 file in a fully consistent manner to generate average energies that are in good agreement with the average decay-type energies. As with Sb-127, two of the gamma-ray transitions are not defined in terms of the originating decay mode (i.e. via β^- decay).

A re-evaluation of the decay data for Te-127 is not required, apart from identifying the decay mode that resulted in all of the gamma-ray transitions.

4.14 52-Te-127m

The decay of Te-127m is dominated by the isomeric transition mode (0.976), with a small β^- component (0.024). All of the decay data in JEF-2.2 are comprehensive and consistent, including the one gamma-ray transition (88.26keV) for IT decay and five gamma-ray transitions identified with the β^- decay mode. However, two of the gamma rays are not defined as originating from this decay mode (both the 593.3 and 658.9keV transitions do occur as a consequence of β^- decay).

A re-evaluation of the decay-scheme data for Te-127m is not required, apart from identifying the decay mode for those gamma-ray transitions not specified in this manner.

4.15 53-I-131

I-131 decays to the ground and metastable states of Xe-131, with the emission of 6 beta particles and 18 gamma rays. All of the data in the JEF-2.2 file are consistent, and the average energies are in good agreement. Furthermore, internal conversion coefficients have been recommended for the major gamma-ray transitions (particularly, the 80.18, 284.3, 364.49, 636.98 and 722.91keV emissions).

A re-evaluation of the decay-scheme data for I-131 is not required. Although the data within the JEF-2.2 file originate from an evaluation in 1983, subsequent measurements have tended to support earlier studies and the original recommendations.

4.16 53-I-132

I-132 undergoes β^- decay to Xe-132, with a Q(beta)-value of 3580(20)keV. A complex decay scheme can be derived from the various measurements, which consists of 34 beta particles and 155 gamma rays. The JEF-2.2 file is reasonably consistent, and is derived from a combination of 33 beta particles and 119 gamma rays. While the spin and parity assignments of I-132 are +0.0, the values adopted in the most recent publications are +4.0 to provide a more suitable basis for defining all of the beta-particle transitions as allowed.

Although the arguments for and against the re-evaluation of the decay data for I-132 are evenly balanced, there are a number of adjustments that should be made. Thus, the gamma-ray data need to be extended, and their introduction will modify the beta-particle emission probabilities; the spin and parity values should also be corrected, and the database could be extended to introduce additional internal conversion coefficients that are not included. On the basis of the above, the decay data for I-132 should be comprehensively re-evaluated.

4.17 52-Te-132

The decay scheme for Te-132 is a simple combination of one beta-particle emission to the 277.86keV nuclear level of I-132 and four subsequent gamma rays. All of these data and the internal conversion coefficients of the gamma-ray transitions are included in the JEF-2.2 file to give a consistent decay scheme.

A comprehensive re-evaluation of the decay data for Te-132 is not required. However, a weighted mean half-life should be calculated to replace the present value in the JEF-2.2 file of 3.258(33) days.

4.18 53-I-138

I-138 undergoes β^- and β^-n decay (branching fractions of 0.945 and 0.055 respectively) to Xe-138 and Xe-137. A complex decay scheme can be derived from the published data which consists of 28 beta particles and 97 gamma rays; all of these data are included in the JEF-2.2 file. However, there are some difficulties in understanding the validity of the continuous spectral data for the gamma-ray and beta-particle decay modes. These specific data have been directly obtained from a private communication of Rudstam et al (1989) to the NEA Data Bank. Furthermore, the continuous spectrum normalisation factors for both of these data sets are zero, which results effectively in the assignment of zero intensity to the spectra when they are extracted from the JEF-2.2 file in a logical manner. Finally, despite the importance of the delayed neutron emissions in the β^-n decay mode of I-138, these data are not included in the file.

Significant efforts are required to produce a complete and consistent decay scheme for I-138. Any such re-evaluation needs to focus on recommending a delayed neutron spectrum, and validating the continuum spectra of the gamma-ray and beta-decay components.

4.19 58-Ce-141

A simple decay scheme can be derived for Ce-141 which consists of two beta-particle emissions and one gamma-ray transition. These data are included in the JEF-2.2 file, along with internal conversion coefficients for the 145.44keV gamma ray. The combination of recommended decay data are consistent, and a re-evaluation is not required.

4.20 58-Ce-144

Ce-144 undergoes β^- decay to the ground and metastable states of Pr-144, with the emission of 3 beta particles and 6 gamma rays. All of these data are included in the JEF-2.2 file, along with internal conversions coefficients for the gamma-ray transitions. The recommended data set is comprehensive and consistent, and a re-evaluation is judged to be unnecessary.

4.21 59-Pr-143

Pr-143 decays to Nd-143, with the emission of two beta particles (a major beta emission directly to the ground state, and a minor transition to the first excited state) and a low-intensity gamma ray. These data are included in the JEF-2.2 file for Pr-143. As expected from the proposed decay scheme, the gamma-ray contribution is negligible (approximately 8.2 x 10^{-3}eV); however, an average gamma (electromagnetic) energy of 310eV has been separately incorporated into the file from another source.

Although of minor importance, there are a number of inconsistencies between the average energies used in the JEF-2.2 that could easily be resolved by re-evaluating the decay data.

4.22 59-Pr-144

Pr-144 (half life of 17.28(5)min) undergoes β^- decay to Nd-144 via the emission of 10 beta particles and 17 gamma-ray transitions; Pr-144m (half-life of 7.2(3)min) decays predominantly by isomeric transition (and one gamma ray), although there is also a small beta branch which includes the emission of 5 low-intensity beta particles and 5 gamma rays. There are a number of deliberate omissions in the JEF-2.2 decay-data file for Pr-144 (only 6 beta particles and 8 gamma rays are included).

While there are no significant inconsistencies in the decay-scheme data for Pr-144 (and Pr-144m), these data should be re-evaluated to produce updated and more comprehensive datasets.

4.23 65-Tb-161

Tb-161 undergoes β^- decay to Dy-161, with the emission of 9 beta particles and 37 gamma rays. However, the JEF-2.2 file contains only the 7 major beta particles and 27 gamma rays (including internal conversion coefficients for the low-energy transitions $\leq$106.15keV).

While the various omissions are only the low-intensity transitions and there are no significant inconsistencies in the recommended decay data, Tb-161 should be re-evaluated to produce an updated and comprehensive dataset.

4.24 35-Br-88

Br-88 has a spin and parity assignment of either -1.0 or -2.0, and undergoes β^- and β^-n decay (0.936 and 0.064 respectively). A relatively complex decay scheme can be constructed from a combination of 59 beta-particle transitions and 166 gamma rays. Overall, the JEF-2.2 datafile appears to be extremely comprehensive, apart from a lack of neutron data.

The average beta (light particle) energy is 1680 keV compared with an average beta-ray decay energy of 2570 keV, while the average gamma (electromagnetic) energy has a value of 4290 keV compared with an average gamma-ray energy of 3260 keV. Although $\overline{E}_\beta$ and $\overline{E}_\gamma$ can be classified as inconsistent in JEF-2.2, some of these differences are not unreasonable since the neutrino component is not included in the average beta-ray decay energy. However, the difference in the gamma data imply that there is a serious inconsistencies between the recommended gamma-ray emissions and the average energy recommended by Rudstam et al (as noted in the comments section of the datafile). It is assumed that the continuous spectra in the gamma and beta fields represent gamma-ray and beta-particle emissions respectively (although this is not clear from the data listings). Apart from this difficulty of identification, both fields include continuum spectrum normalisation factors of zero (i.e. incorrect values, because the resulting spectral intensities would subsequently be calculated as zero).

A detailed re-evaluation is required which would focus on the delayed neutron component, continuum spectra and their normalisation factors and uncertainties. Further measurements are also merited to resolve the inconsistencies in the gamma-ray data.

4.25 35-Br-89

Br-89 has a spin and parity assignment of either -1.5 or -2.5, and decays via the β^- and β^-n modes to Kr-89 and Kr-88 respectively. A Q(beta)-value of 8140 keV was adopted, although a systematic calculation would suggest that a value of 7700 keV is more suitable. Furthermore, a Q(beta)-value of 7426 keV is quoted in the 451-field, which represents the sum of a series of calculations of the various components of the light-particle decay. There are significant inconsistencies in these data and the average energy components (thus, the average beta energy is 2180 keV, while the average beta-decay energy is defined as 2651 keV (higher value)).

There are a relatively large number of gamma-ray emissions (92) that represent a comprehensive database. However, some of these data would benefit from the incorporation of internal conversion coefficients (particularly the 28.51 keV gamma-ray transition). Delayed neutron spectra are not included in the file, and (as with Br-88) the continuum spectrum normalisation factors and their uncertainties are zero in the gamma and beta fields (which is judged to be incorrect).

A re-evaluation of the decay data for Br-89 is recommended to assist in the resolution of the inconsistencies outlined above.

4.26 35-Br-90

Br-90 undergoes β^- and β^-n decay to Rb-90 and Rb-89 respectively. While a decay scheme containing 48 gamma rays can be constructed, only eight gamma-ray emissions have been included in the JEF-2.2 datafile. There are serious inconsistencies in the data that could be resolved by undertaking a more comprehensive re-evaluation. Thus, only two beta particles are postulated in the file with absolute intensities of 10^{-9} (8636.3 and 92.91.7 keV), whereas as many as 35 transitions summing to 100% can be assigned on the basis of the gamma-ray data alone. Furthermore, as with Br-88 and Br-89, the continuum spectrum normalisation factors in the gamma and beta fields are zero (judged to be incorrect).

An extensive re-evaluation is recommended for the decay data of Br-90. Such a study should incorporate a delayed neutron spectrum, all of the observed gamma-ray and beta-particle transitions, and provide non-zero values for the continuum normalisation factors and their uncertainties.

4.27 37-Rb-94

Rb-94 undergoes β^- and β^-n decay to Sr-94 and Sr-93 (0.896 and 0.104 respectively). A relatively complex decay scheme can be derived from the various gamma-ray studies, and these data have been incorporated into the JEF-2.2 file (61 beta-particle transitions and 150 gamma rays). However, there are a number of omissions and anomalies, including a discrepancy between the average beta (light particle) energy of 2830keV and the average beta-ray energy of 3080keV, no delayed neutron spectrum, and the continuous spectrum normalisation factors and their uncertainties assigned values of zero for the gamma-ray and beta decay components, and a lack of internal conversion coefficient data for the low-energy gamma rays.

A comprehensive re-evaluation appears to be necessary, including the incorporation of delayed neutron data, and the introduction of suitable values for the continuum spectra normalisation factors and uncertainties.

4.28 39-Y-98m

No isomeric transition decay has been observed for Y-98m, which is believed to undergo $\beta^-$and β^-n decay (branching fractions of 0.966 and 0.034 respectively). The nuclear level energy is not known with any confidence, although a value of 940keV has been suggested combined with a Q(beta)-value of 9780(200)keV.

While the JEF-2.2 decay data file contains only six beta-particle transitions and 12 gamma-ray emissions, recent measurements have resulted in the assignment of eleven beta particles and 24 gamma rays. Furthermore, there are no delayed neutron data or any internal conversion coefficients in the JEF file.

A new evaluation of the decay data for Y-98m is recommended to bring the data file up to date. Efforts are required to generate delayed neutron data with a realistic normalisation factor for the continuum spectrum.

29 53-I-137

I-137 undergoes β^- and β^-n decay to I-137 and I-136 (0.936 and 0.064 respectively), with a Q(beta)-value of 5877keV. An extremely complex decay scheme can be derived from the gamma-ray measurements, including 108 beta particles and 250 gamma-ray transitions. These data are included in the JEF-2.2 file, including delayed gamma-ray and beta-particle emissions. As with the other nuclides assessed in this exercise, the continuous spectrum normalisation factors and their uncertainties have been erroneously assigned values of zero.

A comprehensive re-evaluation does appear to be necessary, including the addition of delayed neutron data and the recommendation of suitable values for the continuous spectrum normalisation factors.

4.30 30-Y-99

Y-99 undergoes β^- and β^-n decay (0.981 and 0.019 respectively), with the emission of 117 gamma rays and 31 beta particles. Spin and parity assignments of +2.5 have been proposed compared with data-file values of -0.5. Comprehensive new measurements have been carried out since the evaluation undertaken for the JEF-2.2 data file in 1983. Thus, the data file contains only sixteen gamma rays (with zero internal conversion coefficients) and eight discrete beta particles. Furthermore, the normalisation factors and their uncertainties for the continuum spectra of the gamma rays and beta particles are incorrectly assigned values of zero.

An extensive evaluation is recommended for the decay data of Y-99. Such a comprehensive study should include all of the observed gamma rays and the associated beta-particle emissions, as well as delayed neutron data and non-zero values for the continuum normalisation factors and their uncertainties.

4.31 51-Sb-135

The measured decay data for Sb-135 are extremely sparse. Only the half-life, spin-parity (+3.5) and total neutron emission probability (of 0.164(18)) are known with any confidence. Although two gamma-ray emissions have been assigned to this radionuclide (energies of 3292 and 3406keV), there are insufficient data to develop an acceptable decay scheme. Continuum spectrum normalisation factors of 1.00 have been adopted for the gamma-ray and beta spectra, which is viewed as more acceptable than the zero values adopted for the other radionuclides measured above (although their validity requires demonstration).

An evaluation of the available decay data is recommended, including delayed neutron data and specific minor adjustments (e.g. spin and parity of +3.5 (rather than "unknown"), and beta-particle transition(s) and emission probabilities based on a single systematic assessment of the likely population of the Te-135 nuclear level(s)).

4.32 53-I-139

I-139 undergoes β^- and β^-n decay (branching fractions of 0.901 and 0.099 respectively) with a half-life of 2.29 sec. While a significant number of gamma-ray emissions (approximately 100) have been identified with this radionuclide, many of these transitions (52) have not been placed in the nuclear level decay scheme. Furthermore, the average energy components are inconsistent, and extremely low (effectively zero) beta-particle emissions have been adopted of minimal contribution to the proposed decay scheme to give an average beta-particle energy of zero. As with other β^-n decay data, the continuum spectrum normalisation factors and their uncertainties have been erroneously assigned values of zero. Internal conversion coefficients have been adopted for the 22.8, 31.7 and 84.8keV gamma-ray transitions to generate a number of discrete conversion electrons and x-ray data.

A detailed re-evaluation of the decay data for I-139 is recommended, including the introduction of the delayed neutron spectrum. Efforts should also be made to incorporate a significant number of the measured gamma-ray transitions into the proposed decay scheme so that a consistent set of beta-particle data can be generated. The continuum spectral data in the JEF-2.2 file for the gamma and beta components are reasonably comprehensive, although estimates for the normalisation factors and uncertainties are required.

4.33 35-Br-87

Br-87 undergoes β^- and β^-n decay (branching fractions of 0.9743 and 0.0257 respectively), with the emission of approximately 160 beta particles and 360 gamma rays. While the majority of these data are included in the JEF-2.2 file, no beta-particle transitions are included with an energy less than 1810keV (at least 61 emissions), there is no delayed neutron spectrum, the continuum spectrum normalisation factor and uncertainties for the gamma and beta fields are incorrectly assigned values of zero, and there is a lack of internal conversion coefficient data (particularly for the lower-energy gamma rays).

A re-evaluation of the decay data for Br-87 is recommended, including the addition of delayed neutron data and the incorporation of the gamma-ray transitions that have not been placed in the level scheme (approximately 140 of the 360 gamma rays). The addition of the latter would also assist in the assignment of the beta-particle emission, including the lower-energy transitions.

4.34 35-Br-91

The measured decay data for Br-91 are extremely sparse (half-life of 0.541(5) sec). While the branching fractions and Q-values have been evaluated for the β^- and β^-n decay modes (0.817 and 0.183 respectively), no gamma-ray or beta-particle studies have been published to assist in the definition of the decay scheme.

Further measurements are merited to quantify the decay scheme of Br-91 in terms of the beta-particle, delayed neutron and gamma-ray components, although made difficult because of the short half-life. Any form of re-evaluation would be inappropriate until these studies have been undertaken (apart from the addition of the delayed neutron spectrum).

4.35 37-Rb-95

Rb-95 undergoes β^- and β^-n decay (branching fractions of 0.9138 and 0.0862 respectively), with the emission of approximately 55 beta particles and 230 gamma rays. However, the JEF-2.2 data file contains only five beta-particle transitions and twenty gamma-ray emissions; these omissions are also reflected in the major discrepancies between the average beta (light particle) energy and average gamma (electromagnetic) energy compared with the average beta-particle energy and average gamma-ray energy respectively. Although continuous spectra have been generated for the gamma-ray and beta-particle emissions, their normalisation factors and uncertainties have been assigned values of zero; furthermore, there are no delayed neutron data.

An extensive re-evaluation is recommended for the decay data of Rb-95. Such a detailed study should incorporate all of the observed gamma rays, and so derive a comprehensive set of beta-particle transitions, as well as generate a delayed neutron spectrum and non-zero values for the continuum spectrum normalisation factors and uncertainties in the gamma and beta fields.

4.36 37-Rb-93

Rb-93 undergoes β^- and β^-n decay (branching fractions of 0.975 and 0.025 respectively), with the emission of approximately 96 beta particles and 250 gamma rays. A complex decay scheme can be constructed from these data that have been included in the JEF-2.2 file. There are inconsistencies between the average energies taken from the measurements of Rudstam et al and the equivalent data calculated from the individual transitions; furthermore, normalisation factors and uncertainties for the continuous spectra in the gamma and beta fields have been assigned zero values.

A detailed re-evaluation of the decay data does appear to be necessary (particularly the adoption of delayed neutron data and the assignment of valid continuum spectrum normalisation factors and uncertainties).

4.37 33-As-85

The measured decay data of As-85 include the gamma-ray emissions from both the β^- and β^-n decay modes (branching fractions of 0.77 and 0.23 respectively). However, the resulting gamma-ray transitions have not been identified with any of the nuclear levels of daughter Se-85 and Se-84 in JEF-2.2, and a normalisation factor and uncertainty of zero have been adopted for the continuous gamma-ray spectrum.

A re-evaluation of the decay data for As-85 is recommended to produce a reasonably consistent decay scheme, including the introduction of beta-particle emissions that correspond to the observed gama-ray transitions. Delayed neutron data also need to be incorporated into the file, along with non-zero normalisation factors and uncertainties for the continuum spectra.

5 Decay Heat: Relevant Radionuclides

A relatively large number of fission-product radionuclides have been identified in fission yield evaluations as contributing significantly to the decay heat of irradiated fuel (>0.01 of the fractional cumulative yield), although not included in the JEF-2.2 decay-data library. These nuclides are listed in Table 2. However, decay-data files for 33 of these radionuclides can be found in the US ENDF/B-VI library(5) - these files have been assessed to determine their suitability for adoption in the JEF database.

There are no decay-data files to be found in the ENDF/B-VI library 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.

All of the radionuclides have short half-lives (less than 3 sec) taken from the General Electric Chart of the Nuclides assembled by Walker. The decay characteristics of such short-lived nuclides are extremely difficult to quantify in a discrete manner, and therefore these parameters have been calculated in terms of continuum spectra over the energy ranges listed in Table 3. Q-values and branching fractions have been estimated for the β^- and β^-n decay modes (apart from Nd-157 which is defined as undergoing β^- decay only).

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 aim, and a dedicated experimental programme of work would be required to generate discrete spectra and half-life data. Under such circumstances, all theoretical and experimental studies of these individual radionuclides need to be monitored in order to determine when their decay parameters are sufficiently well characterised for a more comprehensive evaluation to be carried out for the JEF or JEFF data library.

6. Concluding Remarks

An initial assessment has been carried out on the JEF-2.2 decay-data files of a list of radionuclides judged to be important by specialists in the European nuclear power industry(9).

Thirty-seven nuclides were singled out for study (see also Table 1):

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

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

c) three radionuclides were found to be reasonably well defined, with the need for only minor modifications.

Work programmes (b) and (c) are considered to be appropriate tasks for financial year 1996/97.

udies were also undertaken of the US ENDF/B-VI decay-data files for a specific set of ..ssion products that contribute >0.01 of the fractional cumulative yield (although not contained within the JEF-2.2 library). These nuclides are some considerable distance from the line of stability, and are very short-lived (see also Table 3). No decay-data files are included in the ENDF/B-VI library for two of the thirty-five nuclides, while the remaining radionuclides involve the adoption of average energies that originate from systematic considerations. These data have been used to generate continuum spectra for the gamma-ray and beta-particle components, while delayed neutron data have been derived from other sources. All papers that describe measurements of the discrete emissions from these radionuclides should be assembled to determine whether the contents of the ENDF/B-VI files can be improved. However, it is generally believed that further experimental studies of these extremely unstable nuclides are required before the ENDF/B-VI decay-data files can be modified with confidence.

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Table 1: Fission Product Nuclides That May Merit Further Evaluation

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

a: UK = United Kingdom, FR = France

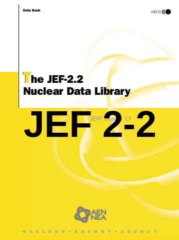

Table 2: Nuclides Missing from JEF-2.2 Decay Data Files that Represents >0.01 of the Fractional Cumulative Yield

Nuclide	JEF-2.2	ENDF/B-VI
39-Y-104	No	Yes
39-Y-105	No	Yes
40-Zr-105*	No	Yes
40-Zr-106*	No	Yes
40-Zr-107	No	Yes
41-Nb-109*	No	Yes
42-Mo-109*	No	Yes
42-Mo-111*	No	Yes
42-Mo-112	No	Yes
43-Tc-113*	No	Yes
43-Tc-114	No	Yes
43-Tc-115	No	Yes
43-Tc-116	No	Yes
44-Ru-115*	No	Yes
44-Ru-116	No	Yes
44-Ru-117	No	Yes
44-Ru-118	No	Yes
44-Ru-119	No	Yes
45-Rh-118*	No	Yes
45-Rh-120	No	Yes
45-Rh-121	No	Yes
46-Pd-121*	No	Yes
51-Sb-141	No	No
57-La-152	No	Yes
58-Ce-153*	No	Yes
58-Ce-154	No	Yes
58-Ce-158	No	No
59-Pr-156	No	Yes
59-Pr-157	No	Yes
60-Nd-157*	No	Yes
60-Nd-158*	No	Yes
60-Nd-159	No	Yes
60-Nd-160	No	Yes
61-Pm-159*	No	Yes
61-Pm-160*	No	Yes

* Nuclide that contributes > 0.1 of the fractional cumulative yield

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Table 3: Partial Summary of ENDF/B-VI Decay Data for Specific Radionuclides

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).