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## Radioactive Heavy Element Decay Data for Reactor Calculations

A.L. Nichols, M.F. James

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# RADIOACTIVE HEAVY ELEMENT DECAY DATA FOR REACTOR CALCULATIONS

A L NICHOLS* and M F JAMES+

SUMMARY

An evaluation has been made of the decay data for 125 heavy element nuclides of interest in nuclear fuel cycle calculations. Computer based data files have been produced in ENDF/B-V format, listing the data references used to formulate the proposed decay schemes and identifying their inadequacies. Evaluated data include half-life, total decay energies, branching ratios, alpha, beta and gamma energies and intensities, spontaneous fission decay data, average energies, internal conversion coefficients and associated uncertainties. Tabulations of the data are presented by means of a variety of computer codes.

*Technology Division, AEE Winfrith
+Reactor Physics Division, AEE Winfrith

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## 1. INTRODUCTION

The United Kingdom Chemical Nuclear Data Committee (UKCNDC) identifies chemical nuclear data requirements for the UK nuclear power programme and coordinates measurement and evaluation programmes to meet specific data requests. One of the committee's specific aims is the production and maintenance of authoritative libraries of nuclear data. These data are required in a form suitable for a wide range of reactor fuel cycle applications, including reactor design and operation, reprocessing, waste management, shielding and transport calculations. The UKCNDC data libraries are maintained and updated periodically, and a summary of their status is published at regular intervals (1, 2).

During 1977 work began to produce a computer-based library of heavy element decay data, UKHEDD-1. The format adopted was ENDF/B-V as recommended for international use (3, 4), permitting the inclusion of spontaneous fission decay data (5, 6). Discussions with scientists at the CEGB Berkeley Nuclear Laboratories produced a comprehensive list of heavy element nuclides requiring decay data evaluation (7), and this was augmented with nuclides from other sources (8, 9, 10) to give the requirements listed in Table 1 and illustrated in Figures 1 and 2. UKHEDD-1 contains comprehensive decay scheme data for 125 nuclides including the reactor fuel actinides, all the principal actinide reaction products up to 253-Es, and their major decay chain nuclides down to 206-Hg. These nuclides are ordered by increasing ZA values (= 1000 x atomic number + mass number), with ground states preceding metastable states, and sequential material numbers from 6500 to 6624.

The IAEA First Advisory Group Meeting on Transactinium Isotope Nuclear Data (11) recommended that new evaluations of the existing decay data be made, including realistic estimates of the data uncertainties. The UKCNDC heavy element decay data library meets these specific requirements. Developments in the construction of this library have been reported annually (12 - 14) to members of the IAEA Co-ordinated Research Programme on the Measurement and Evaluation of Transactinium Isotope Decay Data, and this work has aided in the IAEA compilation of recommended decay data.

## 2. DATA SOURCES

The evaluation effort required data for the following parameters:

(i) half-life;

(ii) Q-values;

(iii) branching ratios;

(iv) alpha energies and intensities;

(v) beta energies, intensities and transition types;

(vi) gamma energies, intensities and internal conversion coefficients;

(vii) spontaneous fission data including prompt neutron and gamma spectra.

All data include associated uncertainties except for the continuous spectra of the prompt neutrons and photons, although uncertainties are given for the average energies of these spectra. Other data included in UKHEDD-1 (mean energies, discrete electrons, X-rays) are derived from the above data using the processing code COGEND (15, 16). The component contributions to the average energies are calculated from the evaluated input data using this code, which has data libraries of fluorescence yields, Auger electron energies, mean X-ray energies, and electron capture ratios. Delayed emissions following spontaneous fission are not included in the library.

The decay scheme of each nuclide has been evaluated from the original references. Every attempt has been made to produce a consistent and comprehensive set of data. References were identified by means of information available through the International Nuclear Structure and Decay Data Network (17) and, furthermore, the literature was surveyed for the most recent publications. The mass tables of Wapstra and Bos (18) were used extensively to determine Q-values, nuclear binding energies and to recalculate radiation energies when resolving inconsistencies.

Conversion electron data are particularly important for the heavy elements, and they have been calculated from the evaluated gamma data and their internal conversion coefficients. When necessary the theoretical internal conversion coefficient tabulations of Hager and Seltzer (19) and Rosel et al (20) have been used.

## 3. EVALUATION PROCEDURE

The published decay data for a nuclide vary in quality and this poses various problems when undertaking an evaluation. Our evaluation philosophy has been outlined in a previous report (21), and can be reviewed in terms of the contents of UKHEDD-1. In general the complexity of the decay data of all the heavy element nuclides, except those that are extremely short-lived, made it advantageous to construct an initial decay scheme from the various data sources. If necessary, weighted adjustments were made to this decay scheme in order to achieve a reasonable degree of consistency, whilst at the same time monitoring any changes that such modifications might have upon the proposed decay data. In this way the decay scheme could be cycled through a series of adjustments whilst maintaining satisfactory agreement with the measured data coupled to specific theoretical data (Q-values and internal conversion coefficients).

For a significant number of nuclides one or two experimental studies stood out as being more detailed and complete than previous measurements. This situation proved commonplace when evaluating gamma data, and wholescale adoption of the work from one particular laboratory was the most realistic procedure.

Beta decaying nuclides proved to be particularly troublesome (for example, 234-Pa and 234m-Pa) because of the lack of beta decay data. In the majority of cases the beta transition data had to be calculated from the gamma data, despite the generally accepted doubts associated with this procedure (22).

Transitions have been introduced into a decay scheme to ensure that there is as much intensity depopulating an excited state as there is feeding it, although these transitions have not been observed experimentally. Frequently supporting evidence for the existence of missing gamma transitions was obtained from either the relevant charged particle reaction studies or the equivalent data of another nuclide decaying to the same daughter nuclear levels.

The metastable and ground states of 246-Am have not been unambiguously identified: 246m-Am has been arbitrarily defined as the product of 246-Pu beta decay, whilst 246-Am has been included in UKHEDD-1 simply for completeness. A small number of metastable nuclides have also been included that are not produced by transmutation or decay in a nuclear reactor and they should be omitted in any reactor-related calculations. These nuclides are 206m-Tl, 207m-Tl, 210m-Bi, 212m-Bi, 212n-Bi, 211m-Po and 212m-Po.

Decay scheme data for 209-Po, formed via the 210-Po (n, 2n) 209-Po nuclear reaction, and its decay product 205-Pb are also included.

Any serious problems encountered during an evaluation have been described in the comments section associated with each nuclide in the library. If the evaluated data contain outstanding problems, a statement has been made to the effect that better measurements are required. However, in the majority of such cases the requirements are comparatively minor and it is unlikely that improvements in the measured data would be of any major importance in nuclear fuel cycle applications.

Every effort has been made to determine a consistent, complete decay scheme and to produce an evaluated data set that can be used with confidence. The consistency of the approved data have been determined by calculating the percentage deviation between the effective Q-value and the calculated Q-value. The effective Q-value is the weighted sum of the evaluated Q-values of the nuclide. The calculated Q-value is the sum of the individual decay components ($\beta^-$, $\beta^+$, $\alpha$ etc) which constitute the total decay. Percentage deviations above 5% are regarded as high and indicate a poorly defined decay scheme. A value less than 5% indicates the construction of a reasonably consistent decay scheme; however, it should be noted that a detailed study of the decay properties may still be lacking because of low specific activity and/or unavailability of the desired material.

## 4. THE HEAVY ELEMENT DECAY DATA LIBRARY, UKHEDD-1

The computer-based library is stored in ENDF/B-V format as defined by Garber et al (23), Kinsey (24) and Tobias (25), based upon standard 80 column card images. There is a general information/descriptive data section (MT = 451, MF = 1) for each nuclide. This section contains the following information:

(i) the name of the evaluator(s) and the date of the evaluation (month and year);

(ii) a list of references used to determine the evaluated data set;

(iii) detailed comments associated with the evaluation;

(iv) various decay data not contained in the decay data section, including beta transition data and specific decay energies;

(v) a consistency check of the evaluated data.

Much of the extra data is obtained from the computer code COGEND (15, 16).

The recommended radioactive decay data are contained within the major data section (MT = 457, MF = 8). The energy data contained within this section are in eV and the intensities are expressed as percentages, calculated from the spectral normalisation factor and the listed relative intensities. The data are listed in the following order in the main section of Appendix 1:

(i) spin and parity of the decaying nuclide;

(ii) half-life;

(iii) average energy per disintegration for light particles, electromagnetic radiations and heavy particles;

(iv) decay modes, Q-values and branching ratios;

(v) radiation decay data, including gamma, beta, alpha, neutron, discrete electron and mean X-ray transitions, and their average energies (the average alpha decay energy includes the recoil energy);

(vi) spontaneous fission decay data.

Much of the data contained in UKHEDD-1, including individual gamma, beta, alpha, discrete electron and X-ray transitions, require no further explanation. However, specific features of the library need to be defined, including the average energy data and the procedures adopted to produce the spontaneous fission decay data.

## 4.1 AVERAGE LIGHT PARTICLE ENERGY

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

$$\overline{E}_\beta = \sum_i^{\text{all }\beta^-} \overline{E}_{\beta_i^-} I_{\beta_i^-} + \sum_j^{\text{all }\beta^+} \overline{E}_{\beta_j^+} I_{\beta_j^+} + \sum_k^{\text{all Auger}} \overline{E}_{A_k} I_{A_k} + \sum_l^{\text{conv}} \overline{E}_{c_l} I_{c_l}$$

where $\overline{E}_{\beta_i^-}$, $\overline{E}_{\beta_j^+}$, $\overline{E}_{A_k}$, and $\overline{E}_{c_l}$ are the mean negatron, positron, Auger electron, and conversion electron energies of the i, j, k, and lth 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. The mean beta energies ($\overline{E}_{\beta_i^-}$ and $\overline{E}_{\beta_j^+}$) are calculated as described by Tobias (26).

## 4.2 AVERAGE ELECTROMAGNETIC RADIATION ENERGY

All electromagnetic radiations such as gamma rays, X-rays, annihilation radiation, and internal bremsstrahlung are included:

$$\overline{E}_\gamma = \sum_i^{\text{all }\gamma} \overline{E}_{\gamma_i} I_{\gamma_i} + \sum_j^{\text{all X-rays}} \overline{E}_{X_j} I_{X_j} + \sum_k^{\text{all }\beta^+} 1.022\, I_{\beta_k^+} + \sum_l^{\text{all }\beta^+\,\beta^-} \overline{E}_{\beta_l} I_{\beta_l}$$

where $\overline{E}_{\gamma_i}$ and $\overline{E}_{X_j}$ are the mean gamma and X-ray energies of the i and jth transition of each type respectively, $I_{\gamma_i}$ and $I_{X_j}$ are the corresponding fractional intensities per disintegration ($I_{\gamma_i}$ is the unconverted (observed) photon intensity), $\bar{E}_{\beta_l}$ is the mean internal bremsstrahlung energy of beta transition l which has an absolute fractional intensity $I_{\beta_l}$, and $I_{\beta^+_k}$ is the absolute fractional intensity of positron transition k. Note that the kinetic energy of any positron decay is already included in $\bar{E}_\beta$, and it is assumed in $\bar{E}_\gamma$ that the positron annihilation occurs at rest.

## 4.3 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 $\bar{E}_{\alpha_i}$, $\bar{E}_{R_j}$, $\bar{E}_{p_k}$, $\bar{E}_{n_l}$, and $\bar{E}_{F_m}$ are the mean alpha, recoil nucleus, proton, neutron and fission fragment energies of the i, j, k, l and mth 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.

## 4.4 SPONTANEOUS FISSION DATA

### 4.4.1 Prompt Energy Release

The prompt energy release from spontaneous fission ($Q_{sf}$) is defined as:

$Q_{sf}$ = (Average kinetic energy of fission fragments after neutron emission)

\+

(Average prompt gamma ray energy)

\+

(Average prompt neutron kinetic energy)

$= Q_{tot} - E_d$

where $Q_{tot}$ is the total energy released and $E_d$ is the average energy released in the radioactive decay of the fission products; that is the sum of the average kinetic energies of beta particles, antineutrinos, gamma rays, delayed neutrons, and recoil nuclei.

$Q_{tot}$ is estimated by the method described in references (27, 28). If m(A, Z) is the mass excess in MeV of the fissioning nucleus, and $y_i$ and $m(A_i, Z_i)$ are the yield and mass excess of the stable fission product $(A_i, Z_i)$, then:

$$Q_{tot} = m(A, Z) - \sum y_i \, m(A_i, Z_i) - \bar{\nu}_p \, m_n$$

where $\bar{\nu}_p$ is the average number of prompt neutrons emitted per fission, and $m_n$ is the neutron mass excess (8.071431 MeV). This equation can only by used as it stands for 252-Cf, as this is the only spontaneously-fissioning nucleus for which a complete set of yields is available. The mass excesses are from reference (18).

For other nuclides an interpolative method has to be used (28). The equation for $Q_{tot}$ may be re-written:

$$Q_{tot} = m(A, Z) - \bar{m}_L - \bar{m}_H - \bar{\nu}_p \, m_n$$

where $\bar{m}_L$, $\bar{m}_H$ are the mean mass excesses of the light and heavy mass peaks. By using available fission yield sets, the approximate dependences of $\bar{m}_L$ on $\bar{A}_L$ (the mean mass number of the light mass peak) and of $\bar{m}_H$ on $\bar{A}_H$ (the mean mass number of the heavy mass peak) can be found (28). For each spontaneously fissioning nuclide suitable values of $\bar{m}_L$ and $\bar{m}_H$ can then be estimated by interpolation.

Extrapolation formulae (27, 28) are used to estimate $E_d$, the fission product decay energy: for consistency these are based on a value of $E_d$ = 23.8 MeV/f for 235-U (n, f).

### 4.4.2 Average Number of Prompt Neutrons

Most experimental values for the average number of prompt neutrons ($\bar{\nu}_p$) are obtained from the review of Manero and Konshin (29). Some of the data have been re-evaluated and these are noted in the reference list of the data file.

For five nuclides (230-Th, 234-U, 239-Pu, 242m-Am and 243-Am) there are no direct experimental data. However, measurements have been made of $\bar{\nu}_{th}$ (A-1, Z), the average number of neutrons emitted following the neutron induced fission of the isotope with mass number one less, so that the same fissioning compound nucleus is formed. For these five nuclides an equation from reference (29) is used to calculate $\bar{\nu}_p$:

$$\bar{\nu}_{th} \, (A-1, Z) = \bar{\nu}_p + 0.101 \, (BE)_n$$

where $(BE)_n$ is the binding energy of the neutron.

For four nuclides (232-Th, 232-U, 235-U and 241-Am) a systematic approach has been used: when A is plotted against

$$\bar{\nu}_p \, (A, Z) - 0.104 \, (Z-90)$$

a reasonably smooth curve is obtained from which $\bar{\nu}_p$ values can be derived.

#### 4.4.3 Prompt Neutron Energy Distribution

The prompt neutron energy distribution is assumed to be Maxwellian, with a mean energy B related to $\bar{\nu}_p$ by Terrell's formula (30):

$$B = a + b\ (\bar{\nu}_p + 1)^{\frac{1}{2}}\ \text{MeV}$$

where a = 0.74 ± 0.02 MeV, and b = 0.653 MeV. An uncertainty of ± 0.020 MeV has been assigned to the value of b by the authors.

#### 4.4.4 Prompt Gamma Energy Distribution

The prompt gamma distributions for all the spontaneous fissioning nuclides are assumed to be an average of the spectra measured for the neutron induced fission of 235-U and 239-Pu and for the spontaneous fission of 252-Cf (31).

## 5. ORDERED TABULATIONS FROM UKHEDD-1

Suitable data listings can be obtained from the ENDF/B-V format data files by means of a number of computer codes developed at the Brookhaven National Laboratory Data Centre and the CEGB Berkeley Nuclear Laboratories. Complete listings of UKHEDD-1 can be obtained as well as selected tabulations of specific decay parameters.

### 5.1 COMPREHENSIVE LISTING OF DATA

Table 1.1 of Appendix 1 summarises the main evaluated decay data in UKHEDD-1, and this is followed by a complete ordered listing of the data produced by processing the library through an interpretation program written at the Brookhaven National Laboratory Data Centre (32). The code LISTFC has been modified at the CEGB Berkeley Nuclear Laboratories and has been used to produce the data listings outlined in Section 4.

### 5.2 SELECTED RADIATION TABULATIONS

Computer codes have been written that can interrogate the library to produce energy ordered listings of alpha, gamma and X-ray data (33). Examples of these tabulations are given in Appendix 2 which consists of the following:

(i) a complete listing of the alpha data by nuclide in ascending order of Z and A;

(ii) a complete listing of the alpha data ordered by increasing energy;

(iii) a complete listing of the gamma and mean X-ray data by nuclide in ascending order of Z and A;

(iv) a complete listing of the gamma and mean X-ray data ordered by increasing energy.

The tabulations of data ordered by increasing transition energy should be particularly useful as aids in the identification of radionuclides that are most readily detected by measuring their characteristic radiation emissions. Also included are the transition intensities, the nuclide identification, the radioactive half-life, and the energy and intensity of the most intense radiations associated with the nuclide. Similar tabulations of other radiations (discrete electrons/beta decay data) can be obtained, if required.

## 6. CONCLUDING REMARKS

If the data library is required for comprehensive reactor-related calculations, the user will require a computer-based copy of UKHEDD-1 (magnetic tape) and some knowledge of the ENDF/B-V format. Enquiries concerning this library and the format should be addressed to:

Mr B S J Davies,
CEGB Berkeley Nuclear Laboratories,
Berkeley, Gloucestershire, England.

Notification of any errors in the library would be welcomed and should be directed to the authors of this report. The intention is to compare UKHEDD-1 with equivalent libraries that are being constructed elsewhere as discussed in (21). In this way the library will be maintained and improved at regular intervals.

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# TABLE 1

## UKHEDD-1 LIBRARY OF HEAVY ELEMENT DECAY DATA

| Nuclide | Material Number | Nuclide | Material Number | Nuclide | Material Number |
|---|---|---|---|---|---|
| 80-Hg-206 | 6500 | 86-Rn-222 | 6542 | 93-Np-240m | 6584 |
| 81-Tl-206 | 6501 | 87-Fr-221 | 6543 | 93-Np-241 | 6585 |
| 81-Tl-206m* | 6502 | 87-Fr-223 | 6544 | 94-Pu-236 | 6586 |
| 81-Tl-207 | 6503 | 88-Ra-223 | 6545 | 94-Pu-237 | 6587 |
| 81-Tl-207m* | 6504 | 88-Ra-224 | 6546 | 94-Pu-238 | 6588 |
| 81-Tl-208 | 6505 | 88-Ra-225 | 6547 | 94-Pu-239 | 6589 |
| 81-Tl-209 | 6506 | 88-Ra-226 | 6548 | 94-Pu-240 | 6590 |
| 81-Tl-210 | 6507 | 88-Ra-228 | 6549 | 94-Pu-241 | 6591 |
| 82-Pb-205 | 6508 | 89-Ac-225 | 6550 | 94-Pu-242 | 6592 |
| 82-Pb-209 | 6509 | 89-Ac-227 | 6551 | 94-Pu-243 | 6593 |
| 82-Pb-210 | 6510 | 89-Ac-228 | 6552 | 94-Pu-244 | 6594 |
| 82-Pb-211 | 6511 | 90-Th-227 | 6553 | 94-Pu-245 | 6595 |
| 82-Pb-212 | 6512 | 90-Th-228 | 6554 | 94-Pu-246 | 6596 |
| 82-Pb-214 | 6513 | 90-Th-229 | 6555 | 95-Am-240 | 6597 |
| 83-Bi-210 | 6514 | 90-Th-230 | 6556 | 95-Am-241 | 6598 |
| 83-Bi-210m* | 6515 | 90-Th-231 | 6557 | 95-Am-242 | 6599 |
| 83-Bi-211 | 6516 | 90-Th-232 | 6558 | 95-Am-242m | 6600 |
| 83-Bi-212 | 6517 | 90-Th-233 | 6559 | 95-Am-243 | 6601 |
| 83-Bi-212m* | 6518 | 90-Th-234 | 6560 | 95-Am-244 | 6602 |
| 83-Bi-212n* | 6519 | 90-Th-235 | 6561 | 95-Am-244m | 6603 |
| 83-Bi-213 | 6520 | 91-Pa-231 | 6562 | 95-Am-245 | 6604 |
| 83-Bi-214 | 6521 | 91-Pa-232 | 6563 | 95-Am-246* | 6605 |
| 83-Bi-215 | 6522 | 91-Pa-233 | 6564 | 95-Am-246m | 6606 |
| 84-Po-209 | 6523 | 91-Pa-234 | 6565 | 96-Cm-241 | 6607 |
| 84-Po-210 | 6524 | 91-Pa-234m | 6566 | 96-Cm-242 | 6608 |
| 84-Po-211 | 6525 | 91-Pa-235 | 6567 | 96-Cm-243 | 6609 |
| 84-Po-211m* | 6526 | 92-U-232 | 6568 | 96-Cm-244 | 6610 |
| 84-Po-212 | 6527 | 92-U-233 | 6569 | 96-Cm-245 | 6611 |
| 84-Po-212m* | 6528 | 92-U-234 | 6570 | 96-Cm-246 | 6612 |
| 84-Po-213 | 6529 | 92-U-235 | 6571 | 96-Cm-247 | 6613 |
| 84-Po-214 | 6530 | 92-U-235m | 6572 | 96-Cm-248 | 6614 |
| 84-Po-215 | 6531 | 92-U-236 | 6573 | 96-Cm-249 | 6615 |
| 84-Po-216 | 6532 | 92-U-237 | 6574 | 96-Cm-250 | 6616 |
| 84-Po-218 | 6533 | 92-U-238 | 6575 | 97-Bk-249 | 6617 |
| 85-At-215 | 6534 | 92-U-239 | 6576 | 97-Bk-250 | 6618 |
| 85-At-217 | 6535 | 92-U-240 | 6577 | 98-Cf-249 | 6619 |
| 85-At-218 | 6536 | 93-Np-236 | 6578 | 98-Cf-250 | 6620 |
| 85-At-219 | 6537 | 93-Np-236m | 6579 | 98-Cf-251 | 6621 |
| 86-Rn-217 | 6538 | 93-Np-237 | 6580 | 98-Cf-252 | 6622 |
| 86-Rn-218 | 6539 | 93-Np-238 | 6581 | 98-Cf-253 | 6623 |
| 86-Rn-219 | 6540 | 93-Np-239 | 6582 | 99-Es-253 | 6624 |
| 86-Rn-220 | 6541 | 93-Np-240 | 6583 | | |

*These nuclides are neither formed by neutron transmutation nor are they the products of actinide decay; they are included for completeness (see Section 3).