Chapter 9

DECAY HEAT VALIDATION STUDIES

Introduction

An accurate prediction of total decay heat and its time dependence is required in studies of loss of coolant accidents, and also in connection with the transportation and storage of spent fuel. At short cooling times, the main component is due to the decay of the fission products and also to ^{239}U and ^{239}Np decay, which are important for cooling times up to 15 days. In the case of fast reactors the alpha decay of actinide isotopes and the decay of activated steel and ^{24}Na make a significant contribution. The importance of the different contributions has been discussed by A. Tobias [1] and by F. Storrer in JEF/DOC-473 [2]. For shielding studies calculations of the gamma energy spectrum associated with the decay are also required. The gamma emission in beta decay of fission products is also an important component of the total gamma emission in normal reactor operation.

The fission product nuclide densities depend on the yields and decay constants of all the nuclides in a decay chain and to a lesser extent on the cross-sections for transmutation to another isotope. The decay heat also depends on the decay energies of the fission products [22]. For the gamma spectrum calculation the spectra of the gamma emissions of the individual isotopes are required. These data are included for a large number of fission products in JEF-2.2, and also in the ENDF/B-VI and JNDC-FP-V2 [16] evaluated nuclear data files.

Studies have been published in which summation calculations made using JEF-2.2 fission yield and radioactive decay data have been compared with fission product integral decay heat measurements following a fission pulse. In particular the works of F. Storrer, CEA Cadarache (JEF/DOC-441 [3]), the Saclay group of B. Nimal, J. Blachot, J.C. Nimal and C.M. Diop (JEF/DOC-442 [4]) and that of the Delft University group, P. de Leege, J. Hoogenboom and J. Kloosterman [5]. The latter paper also makes comparisons with calculations made using ENDF/B-VI and JNDC-FP-V2 data.

One should mention that because the thermal leakage is very small in the case of a FBR, the integral reactor decay power can be measured by a calorimetric method (e.g. as it was measured in PHENIX, SUPERPHENIX [23] and the JOYO reactor). It is also to be noted that the Saclay group has initiated a project (the MERCI experiment) to apply such a calorimetric measurement to a PWR irradiated fuel element.

We also note the sensitivity and uncertainty studies made by James, *et al*. based upon the FISPIN inventory code and associated UK data library [31], by the Saclay group of C. Diop, B. Nimal, J. Rebah [18] and colleagues using the DOP-699 (CEA-86) database and by the Japanese Decay Heat Working Group [19] using JNDCFP-V2 and ENDF/B-VI data (of interest also to the assessment of the JEF-2.2 data which has elements in common). The Japanese group has also summarised the sources of differences and attempted to identify discrepant or missing isotopic decay data, and studied the effect of using data obtained by means of the gross theory of beta decay [12,29].

F. Storrer has described the development of the JEF-2.2 decay data library and has noted the following stages in the development:

JEF-2.2.0	January 1992
JEF-2.2.1	July 1992
JEF-2.2.2	February 1993
JEF-2.2.3	July 1993

The decay heat calculations by F. Storrer [3] and by B. Nimal, *et al.* [4] predate this final version.

Decay heat standards

It is useful to consider the fission product decay heat following a fission pulse. The measurements of total decay heat (and of the beta and gamma components when a spectroscopic technique is used, rather than a calorimetric one) have been made for a set of finite irradiation times, but these can be unfolded to obtain the heat following a fission pulse. Similarly the fission product decay heat following a finite irradiation period can be calculated from the fission pulse response, by integration (with allowance for any transmutation and delayed neutron effects, etc.). Based on the fission pulse responses obtained by unfolding the measurements, measurements made over different irradiation periods can be compared and can be averaged to obtain a best estimate fission pulse response. The fission pulse response can be fitted by a weighted sum of decaying exponentials. These curves are called elementary fission curves or decay heat burst functions. A. Tobias has analysed the measurements of total decay heat [6], as well as the beta and gamma components, for ^{235}U and ^{239}Pu to obtain best estimate fission pulse responses for decay periods up to 10^5 s. The method used is similar to that employed by Schmittroth and Schenter to derive the American National Standard for Decay Heat Power in Light Water Reactors (the ANS standard) ANS-5.1 [7], which represents the functions by a sum of 23 exponentials. There is a difference, however, because the ANS standard includes in the weighted averaging the results of fission product summation calculations, and for times longer than 10^5 s it is based on the summation calculations alone (no direct measurements being available). (This standard also includes ^{238}U, the curve being based on fission product summation calculations only.) The JAERI standard (for both LWRs and FBRs) [8], JAERI-M-91-034, is based on summation calculations alone. It uses 33 exponentials and treats the isotopes ^{235}U, ^{238}U, ^{239}Pu, ^{240}Pu and ^{241}Pu. This is the only standard treating decay heat in both LWRs and fast reactors. There is also the German standard, DIN 25463 and a draft ISO standard [7].

It is simplest to make comparisons between the summation calculations and one of these standards – Tobias, ANS or JAERI – rather than with the separate measurements.

The comparisons made by F. Storrer between the different standards have been extended by J. Rebah to the case of infinite irradiation for ^{235}U and ^{239}Pu thermal fission. The fission product decay heat released after some typical reactor irradiation cycles was also investigated by Rebah in his PhD thesis, using the DOP-699 library [18]. Yoshida, *et al*. have emphasised the impact of the discrepancy between the calculated and measured gamma-ray component, observed for the case of burst fission, on the calculated value for the one year irradiation cycle [12].

The ANS-5.1 1993 standards for ^{235}U and ^{239}Pu total decay heat differ from the Tobias standards by about ±4%, being larger than the Tobias standards for cooling times greater than about 15 s. The JAERI-M-91-034 standards have larger differences relative to the Tobias standards. The ^{235}U

standard is about 10% lower at 1 sec. and then up to 5% lower for times between 3 and 10^3 s and then becomes, unexpectedly, 5% higher at 10^4 s. This is probably due to the large number of short lived fission products introduced into the Japanese library (and standard) based on theoretical investigations.

The ^{239}Pu standard is also lower than the Tobias standard for times up to about 10^3 s (by up to about 6%). One should keep in mind that, as pointed out by K. Dickens [21], there is a clear and still outstanding discrepancy in the gamma-ray decay heat data for cooling times between 600 and 6 000 s between the different experimental data themselves (LANL calorimetric vs. ORNL spectroscopic data), but also with the summation calculations, leading to a significant underestimation of the gamma decay heat with all the fission product data files. Although Yoshida, *et al*. [12] have given a possible explanation for this discrepancy (suspecting some missing beta feeding in the decay of some fission products with high *Q* values or a problem of assignment between ground and meta-stable state), the wait continues for complementary integral data (for instance from the recent measurements at Lowell University) or for new decay data for individual/separated FPs from spectroscopic investigations.

Decay heat measurements

The measurements which are most often used to assess the accuracy of summation calculations are those made by Dickens, *et al*. [9], by Akiyama and An [10] and by Yarnell and Bendt [11]. The measurements by separated beta and gamma spectroscopy made by Akiyama, *et al*. were in a fast reactor spectrum, but the differences between fast and thermal spectrum effects are not very great because the corresponding fission yield distributions are not so different. Dickens, *et al*. irradiated thin foils of ^{235}U and ^{239}Pu (of about 10 μg) for 1, 10 and 100 s, and measured (also by a spectroscopic method) the decay heat at cooling times between 2 and 2 000 s. Yarnell and Bendt irradiated thin foils of 66 mg of ^{233}U, ^{235}U and ^{239}Pu for 20 000 s and measured, using a cryogenic calorimeter, the total decay heat at cooling times between 10 and 10^5 s after the irradiations.

Among the new available experimental data to be further investigated, the work by K. Deadman, *et al*. (measurements in ZEBRA relating to fission in ^{235}U and ^{239}Pu) [24] should be mentioned, as should that of the Lowell group (using the UML 5-MV Van de Graaff accelerator, fission of ^{235}U, ^{238}U and ^{239}Pu) [25,26].

Relative contributions of the different types of decay data

The study by F. Storrer [3] describes the types of evaluated isotopic decay data included in the evaluations, as defined by Dickens [22]. The JEF-2.2 isotopic decay data can be considered to be of one of three types (see also Chapter 2 which describes the JEF-2.2 radioactive decay data library):

(a) Decay schemes constructed from detailed spectroscopic measurements and average beta and gamma energies derived from the level scheme.

(b) Average beta and gamma energies measured directly (continuous beta and gamma-ray spectral distributions are measured and then integrated). These are the data evaluated by G. Rudstam, *et al*. [3] based on the measurements made on OSIRIS.

(c) Theoretically derived decay data. These are the values derived by H. Klapdor-Kleingrothaus, *et al*. using the proton-neutron quasiparticle random phase approximation.

In a few cases, the Type (a) spectra have been given (in ENDF-6 format) [15] together with the Type (b) data, but with a unique set of average beta and gamma energies corresponding to G. Rudstam's evaluations. It should be noted that this is leading to some ambiguities in the recommended ENDF-6 procedure to use the data and to some problems in the subsequent internal consistency checks of such decay data based on discrete spectral information.

The JEF-2.2 decay data library differs from the JNDC-FP-V2 and the ENDF/B-VI files 29, in that there are no cases where spectra for some short-lived fission products have been augmented by calculated values derived using the gross theory of beta decay.

The Type (b) and (c) evaluations were included in JEF-2.2 because of the much improved results obtained for decay heat predictions when including these new data by G. Rudstam, *et al*. [27] and by H. Klapdor-Kleingrothaus [28].

Considering the total decay heat for a ^{235}U fission pulse calculated with JEF-2.2, F. Storrer shows that for cooling times longer than about 3×10^6 s the calculated heat is determined by the data of Type (a), while for times shorter than 3×10^5 s the largest contribution is from isotopes having data of Type (b). For a cooling time shorter than about 10 s the Type (c) data makes its maximum contribution of about 25%.

A similar study has been made by T.R. England, *et al*. using the ENDF/B-VI library [29].

Comparison of JEF-2.2 summation calculations with the Tobias standards and with measurements

Fission pulse total decay heat

Comparing the JEF-2.2.2 summation calculations with the Tobias best fit to the measurements for ^{235}U total decay heat, F. Storrer shows that the agreement is improved relative to JEF-1 at cooling times shorter than about 100 s. This improvement is attributed to the larger number of short lived fission products treated in JEF-2.2 (mainly from Type (b) evaluations) and to the improved quality of the data. However, an underestimation of the total decay heat of about 6% remains in the cooling interval from about 10 s to about 1 000 s. At 10^4 s there is an overestimation of about 5%.

Compared with the Tobias best fit for ^{239}Pu the summation calculations underestimate in the time interval 10 to 1 000 s by up to about 8%. Above about 10^4 s there is a small overestimation, and below 10 s there is no clear improvement relative to JEF-1.

Beta and gamma components of the fission pulse decay heat

Storrer has compared JEF-2.2.1 summation calculations with the Tobias standards for the beta and gamma components of ^{235}U and ^{239}Pu. The gamma component of the decay heat is underestimated for times shorter than about 3 10^3 s. The underestimation is over 10% for ^{235}U between 10 and 1 000 s and reaches about 15% at 300 s. For the gamma component of ^{239}Pu, there is a similar underestimation but an overestimation exists below 10 s. For cooling times longer than about 3 10^3 s there is a small overestimation for both ^{235}U and ^{239}Pu.

For the ^{235}U beta component there is an underestimation of up to 15% at times shorter than about 30 s and an overestimation of up to 5% between 30 and 2 10^4 (with a relative minimum at 700 s). For ^{239}Pu, the beta component of the Tobias standard is underestimated at cooling times shorter than about 300 s, the underestimation reaching about 13% at 10 s cooling. At 2 000 s, and again at about

$2\ 10^4$ s, there is an overestimation of about 6%. For times less than about 300 s the summation calculation lies between the Tobias standard and the JAERI standard, and it is close to the JAERI standard at times longer than about 10^4 s.

Comparisons with measurements of the ^{239}Pu gamma component of the heating

Yoshida, *et al*. [12] have compared JEF-2.2 calculations of the gamma energy for a ^{239}Pu fission burst with the measurements by Dickens, *et al*., Akiyama and also with calculations made using JNDC-V-2 and ENDF/B-VI. All three data libraries underestimate the gamma energy in the 300 to 3 000 s interval, but for JEF-2.2 the underestimation is larger (about 10%) and extends down to about 30 s. The authors attribute this to the JEF policy of not augmenting measured gamma spectra to take into account theoretical estimates of the total gamma energy yield and that thus the data suffer from the so-called "pandemonium effect". As a consequence it is expected that the beta component of the decay heat in this time interval will be overestimated. Yoshida, *et al*. also identify isotopes for which the differences between the data in JEF-2.2 and in JNDC-V-2 have significant effects in the time interval 300 to 3 000s.

The study by B. Nimal, et al. comparing JEF-2.2.2 summation calculation with the measurements of Dickens, et al., Akiyama, and Yarnell and Bendt.

In JEF/DOC-442, B. Nimal, *et al*. compare JEF-2.2.2 summation calculations with the measurements of Dickens, *et al*., Akiyama, and Yarnell and Bendt [4].

^{235}U thermal fission pulse, beta energy

Between 20 and 200 s cooling time the summation calculations are about 10% higher than the measured values of Dickens, *et al*. and elsewhere are within the range of uncertainties. The Akiyama values (fast reactor spectrum) are higher in this cooling time interval and close to the summation calculations.

^{235}U thermal fission pulse, gamma energy

Between 20 and 100 s cooling time the summation calculations are about 10% lower than the measured values of Dickens, *et al*. and they are about 15% higher at 3 000 s. Elsewhere they are within the range of uncertainties. However, the Akiyama values are higher than the summation calculations at this cooling time and close to them.

^{235}U thermal fission pulse, total decay energy

The summation calculations are within the uncertainties of the measured values of Dickens, *et al*. and Akiyama, although tending to be up to about 5% higher than the Dickens values between 100 and 10^4 s.

^{239}Pu thermal fission pulse, beta energy

Below about 20 s cooling time the summation calculations are up to about 10% lower than the measured values of Dickens, *et al*. and elsewhere are within the range of uncertainties.

^{239}Pu thermal fission pulse, gamma energy

As noted above, based on the study by Yoshida, *et al*. [12], between 30 and 3 000 s cooling time the JEF-2.2 summation calculations are about 10% lower than the measured values of Dickens, *et al*. and Akiyama.

^{239}Pu thermal fission pulse, total decay energy

The summation calculations are lower than the Dickens measured values between 10 and 100 s and at about 1 000 s, although not far outside the uncertainty margins.

Irradiation of 10^{13} s

The ^{235}U beta energy and total decay heat are overestimated by a few per cent, relative to the measurements of Dickens, *et al*. for cooling times of up to about 100 s. There is a similar slight overestimation of the beta component in the case of ^{239}Pu, but there is an underestimation of the gamma component at short cooling times (up to about 300 s) by up to about 8%. The net result is a small underestimation of the total decay heat, compared with the measurements of Dickens, *et al*.

Comparison with the measurement of Yarnell and Bendt for ^{235}U total decay heat (2.10^4 s irradiation)

There is a tendency for the summation calculations to underestimate the measured values between 10 s and 200 s cooling times, but the results are close to the uncertainty ranges.

Gamma spectra for ^{235}U fission at 1 000 s cooling

The summation calculations have been compared with the measurements of Dickens, *et al*. and the agreement appears to be satisfactory.

The study by P. de Leege, J. Hoogenboom and J. Kloosterman

These authors have made a comparison between summation calculations, using the yield and decay data of the JEF-2.2, ENDF/B-VI and the JNDC FP libraries, as well as the measurements of Dickens, *et al*. and Yarnell and Bendt and also with the analytical ANS and JAERI standards. Calculations have been made for ^{233}U, ^{235}U and ^{239}Pu. The calculations were made using the module ORIGEN-S from the SCALE-4.2 code [13], using updated ORIGEN-S libraries from ENDF/B-VI, JEF-2.2, and JNDC FP data respectively. These new data libraries were produced in the Netherlands in a joint effort of the Delft University Interfaculty Reactor Institute (IRI) and the Netherlands Energy Research Foundation (ECN, Petten) [14].

Dickens

The analytical ANS-5.1 and JAERI standards give results which are both about 10-25% higher than the experimental results for 1, 10 and 100 s irradiation times. The JAERI standard is slightly better than the ANS-5.1 standard. The same discrepancies between experimental and summation calculation results are found for the ENDF/B-VI, JEF-2.2, and JNDC evaluated data libraries.

A remarkable discrepancy is found for the long irradiation time (100 s) and long cooling time (5 000 s) for both analytical (standards) and evaluated (summation calculation) results. There is good agreement between analytical and evaluated results. For ^{239}Pu the analytical and certainly the evaluated results are better than the results for the ^{235}U benchmark. The results of the different evaluated data are closer than in the ^{235}U benchmark.

Yarnell and Bendt

For ^{233}U no analytical (standard) data are available from ANS-5.1. The analytical result from JAERI shows a difference ranging from about -5% for short cooling times to about 0% for long cooling times. The evaluated results have discrepancies from about -10 to -5% for JEF-2.2 and ENDF/B-VI data, while the discrepancies for JNDC data ranges from about -5 to 0%. For ^{235}U both the ANS-5.1 and the JAERI analytical results are in good agreement with the experimental results, certainly the ANS-5.1 result. The evaluated results show discrepancies from about -5 to 0% for JNDC and JEF-2.2 data, with a larger discrepancy for the ENDF/B-VI data. For ^{239}Pu the analytical results show a discrepancy of about -5% for ANS 5.1 over the whole decay time, while the JAERI result shows a discrepancy from about -10 to -5% for short to long cooling times. The evaluated results show discrepancies from about -15 to 0% for short to long cooling times. The discrepancies at a cooling time of 10^5 s are about +5 to +10%. The best agreement is given by JNDC and JEF-2.2 evaluated data.

Summary of the Delft University study

In this study it is found that the decay heat is overestimated for both the ^{235}U and ^{239}Pu measurements made by Dickens (short irradiation times) for both analytical and evaluated solutions by about +10 to 25%. There is good agreement between the independent analytical and evaluated decay heat calculation results.

The decay heat is underestimated for all three nuclides studied in the Yarnell and Bendt measurements (long irradiation times) for both analytical and evaluated solutions by about -5 to -10%, except for decay heat from ^{239}Pu at very long cooling times. For long cooling times (after 1 000 s) the agreement between experiments and calculations is rather good.

Studies of uncertainties in decay heat predictions arising from uncertainties in the basic data

J. Rebah [18] has made a very careful analysis of the calculated fission product decay heat uncertainty due to the uncertainty on FP data, i.e. the half-lives, average decay energies and fission yields. These data had previously been treated as independent parameters, but various assumptions of internal correlations were investigated to estimate more realistic final uncertainties, especially for the uncertainty on the average decay energy, strongly dependent upon the distance of the FP from the β stability line (i.e. upon the value of (Zp - Z), Wahl's key parameter for neutron-rich FPs with short half-life and high *Q* value). For this energy parameter, he demonstrated that the assumption of zero correlation is leading to a large underestimation of the resulting uncertainty on decay heat, and full correlation to a large overestimation, whereas a correlation factor based on the (Zp - Z) parameter seems more reasonable. J. Rebah has also compared some elementary fission curves with results from summation calculations made using both the DOP-699 and JEF-2.2 data files for ^{235}U and ^{239}Pu thermal fission (see Figures 6.6 and 6.7 in [18]). It should be noted that the covariance data needed to estimate the internal correlations among the FP data are not available in JEF-2.2.

In Japan systematics are being used to estimate the uncertainty on the average decay energies for the FPs which have been completely calculated using the gross theory of beta decay (but no internal correlation has been taken into account by Oyamatsu, *et al*. [19] in his simplified method to calculate the decay heat uncertainty due to FP data).

One should also note the technical difficulty in providing full covariance information and processing it in a decay data library (lacking in most files except for those dealing with radionuclide metrology) as pointed out by G. Winkler [20].

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