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REVIEW OF DECAY HEAT CALCULATIONS

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Abstract: Calculations of decay heat of fission products were reviewed and compared with recent measurements. The calculated decay heat with known nuclear data alone was considerably smaller than the measured result at short cooling times less than 10^3 s. The estimated decay energies have most significant influences on the decay heat. The theoretical estimation of unknown or imprecisely-known decay energies by the gross theory of beta decay made it possible to reproduce quite well (within ±5%) the decay heat measurements at University of Tokyo (UT) and at Oak Ridge National Laboratory (ORNL) for variety of fissiles. The theoretical estimation of unknown beta decay data by microscopic nuclear structure calculations could also improve the accuracy considerably.

(decay heat, gamma spectrum, fission product, summation calculation, capture effect, uncertaintly, review)

Introduction

The first self-sustaining chain reaction was achieved by Enrico Fermi group at Chicago University in 1942 shortley after the discovery of fission in 1939. Since then many types of reactors have been developed, for example Gas-Cooled Reactor (GCR), Light Water Reactor (LWR), Heavy Water Reactor (HWR), Liquid-Metal-Cooled Fast Breeder Reactor (LMFBR), and so on. Irrespective of which reactor system one may consider, there are a number of important design and operating criteria which require a knowledge of decay heat of fission products, as well as other characteristics. The decay heat of fisssion products (FPs) plays an important role in predicting the heatup of the nuclear fuel rod during a loss-of-coolant accident (LOCA) of a LWR. The decay heat is also important in designing the heat removal system of a reactor and also spent fuel handling equipments.

Decay heat may be determined by either integral measurements on mixed fission products or by calculations. Although the calculation may be preferred, by virtue of its generality, in the earlier years experimental results were required to fill the gaps at short cooling times as decay data for short-lived fission products were sparse to calculate the decay heat accurately at short cooling times.

Extensive works have been done for decay heat measurements and calculations since 1940s and the accuracy has been improved with time both in the measurement and the calculation. There has been considerable improvement, in recent years, in the status of decay heat calculation, that is, in the status of fission product decay data. Nevertheless, it is still necessary to measure decay heat accurately for some typical conditions in order to verify the calculation and thus establish confidence in the calculation method and its results. Therefore, measurements and calculations of the decay heat are considered complementary.

In the present paper the decay heat calculations are reviewed with primary interest in the applicability and the accuracy of the calculation. The calculated results are compared with experimental ones to assess the accuracy. This subject has been reviewed on a number of occasions in the past[1-8], especially in 1979 by Schrock[9] and in 1980 by Tobias[10]. Therefore, in the present paper the works in 1980s are primarily reviewed and the works before 1980 are supplimented by the review of Tobias[10]. The actinide decay heat is not included in the present review but the gamma-ray spectrum of fission products is included as well as beta, gamma, and beta plus gamma decay heats of fission products. Also included are the discussion on uncertainties of the calculation and the effect of neutron capture transformations of fission products.

Review of Decay Heat Calculation and Its Assessment with Experimental Data

Calculation Method and Necessary Nuclear Data

There are two approaches to calculate the decay heat of fission products, the statistical method[11,12] and the summation method. Way and Wigner (1948)[11] considered fission products as a sort of statistical assembly and used empirical relations for both the radioactive half-lives and atomic masses to obtain the gamma (P_γ MeV/fission-s), and beta plus gamma (P_t MeV/fission-s) decay heat following an instantaneous burst of thermal fissions in ^{235}U as follows,

$$P_\gamma = 1.26\ t^{-1.2} \qquad (1)$$

$$P_t = 2.66\ t^{-1.2} \qquad (2)$$

for decay times t (s) in the range 10 s - 100 d. The statistical method used by Way and Wigner was the only method to be able to estimate the decay heat at short cooling times by calculation at the time and the above equations were used extensively for a number of years. The method is also much simpler than the summation calculation, however, it is imcomplete by nature and less accurate at longer cooling times. Therefore, the statistical method was gradually superseded by the summation calculation.

The summation calculation determins, in the first instance, the isotopic concentration of each fission product nuclide produced by the irradiation of fuel by solving the linear system of coupled first order differential equations

which describe the buildup and decay of fission products as follows:

$$\frac{dN_i}{dt} = -(\lambda_i + \sigma_i\phi)N_i + \sum_j f_{j\to i}\,\lambda_j N_j + \sum_k \mu_{k\to i}\,\sigma_k\phi N_k + y_i F, \qquad (3)$$

where
- N_i : concentration of nuclide i ,
- λ_i : decay constant of nuclide i ,
- σ_i : average capture cross section of nuclide i ,
- ϕ : neutron flux
- $f_{j\to i}$: branching ratio of the decay from nuclide j to i ,
- $\mu_{k\to i}$: production rate of nuclide i per one neutron capture reaction of nuclide k,
- y_i : independent fission yield of nuclide i ,
- F : fission rate.

There are two methods to solve coupled differential equations (3), i.e. the matrix method[13,14] and the Bateman method[15-18]. The former solves equations by means of numerical integration and the latter analytically.

Once the inventories of fission products have been obtained for the required conditions of irradiation and cooling, the decay heat produced may be simply derived by summing the nuclide activities with the weights of beta (E_β^i) and gamma (E_γ^i) energy release per disintegration of that nuclide; i.e.

$$P_\beta(t) = \sum_i \lambda_i N_i(t)\, E_\beta^i \qquad (4)$$

$$P_\gamma(t) = \sum_i \lambda_i N_i(t)\, E_\gamma^i \qquad (5)$$

where $P_\beta(t)$ and $P_\gamma(t)$ are beta and gamma decay heat, respectively at a cooling time t after shutdown. Beta or gamma spectrum of fission products can also be calculated easily once the inventories are obtained by changing the weights from the mean decay energy to the energy spectrum of each nuclide for the summation of activities. Therefore, the summation method is, by nature, more general and versatile than the statistical method.

From Eqs.(3) - (5) it is clear that following nuclear data are necessary for each fission product nuclide to obtain the inventory and then the decay heat:

(1) Decay constant,
(2) Decay scheme data including branching ratios,
(3) Decay energies including spectrum data,
(4) Neutron capture cross section data, and
(5) Fission yield data.

Early Works

The first summation calculations of the decay heat following ^{235}U thermal fission would appear to be those of Howlett et al. (1950)[19], which used a nuclear data library of only 84 radioactive nuclides. The decay heat was given in the table for the cooling time range 1 d - 5 y.

All of the early summation calculations suffered from the same problem, i.e. a lack of data on individual nuclide decay for many short-lived fission products known at the time. Consequently, the results of such calculations could be regarded as valid only for times in excess of a few hours. The accuracy at short cooling times was improved slowly with time due to accumulation of nuclear data for short-lived nuclides.

There are two trends to fill the gap at short cooling times. An attempt was made by Stehn and Clancy (1958)[1] to combine the evaluated mean experimental curve at short cooling times with the results of their own summation study, to yield a composite curve of total decay heat following both the burst fissions and an infinite irradiation. This work was to form the basis of a number of decay heat evaluations in the following years. Shure (1961)[2] made an extensive review of previous reviews, existing decay heat measurements and summation calculations. Shure recommended the infinite-irradiation total and gamma decay heat curves based upon the evaluation by Stehn and Clancy (1958)[1] at short cooling times less than 10^3 s and upon summation calculations by Perkins (1963)[20] at longer times. A seven-energy group gamma spectrum was also given by Shure (1961)[2], and this was again based upon measurements at short cooling times less than 10^3 s and upon summation calculations[20] at longer cooling times. The work by Shure was used extensively for a number of years and formed the basis for the proposed 1973 ANS standard[21] for ^{235}U thermal fission, some 10 y later.

The other attempt to fill the gap at short cooling times was based upon the summation calculation alone with an improved nuclear data library where unknown nuclear data for short-lived nuclides were estimated theoretically or statistically. One notable example was the extensive work by Bolomeke and Todd (1957)[22]. They used a nuclear data library which contained data for 223 radioactive nuclides of which unknown data for 89 nuclides were estimated theoretically. It is widely accepted that a turning point in the development of summation calculation was exemplified by the work by Perkins and King (1958)[23]. Predictions were made of seven-group gamma spectra, and beta and gamma decay heat, following ^{235}U thermal fission for shutdown times as short as 100 s. In making these calculations it was necessary to estimate the decay properties of over 20 short-lived nuclides in a data library of 123 fission products.

Works in 1970 s

In 1970 s the summation method for calculating fission product decay heat was rapidly improving in accuracy due primarily to the inclusion of more short-lived fission products in the data library and the improvement in the estimation method of unknown decay data for these nuclides.

The IAEA convened two related meetings in 1973, one is the IAEA Symposium on Applications of Nuclear Data in Science and Thechnology and the other is an Advisory Group Meeting on Fission Product Nuclear Data. Devillers et al.[24] compared results of their summation calculations with the latest calorimetric measurements by Lott et al. (1973)[25] for ^{235}U thermal fission. Lott[4] made a review on decay heat at the latter meeting. Lott compared results of eight different summation studies with each other, with the evaluation by Shure[2], and with a number of experimental results. It was concluded that although the summation studies were consistent with each other, they were not consistent with the experimental values. In addition there were considerable discrepancies between many of the experimental values themselves. Hence the need for further decay heat measurements were emphasized. Lott also suggested

that, in future summation studies, the overall uncertainties in the results should be evaluated in direct relation to those in the basic data.

A number of more detailed calculation of fission product decay heat were made by Tasaka and Sasamoto (1974)[26] using the summation code FP-S[27] with a data library of 443 radioactive nuclides which included a number of unknown fission products, for which nuclear data were estimated. Comparisons were made with many of the experimental results. It was concluded in this study that summation calculations could adequately predict decay heat from fuel which is not irradiated in high thermal neutron fluxes.

Towards the end of 1974 the ENDF/B-IV fission product nuclear data file[28] had been assembled and initially tested by Kee et al.[29] with the summation codes ORIGEN[13] and RIBD-II[30]. It was concluded that, provided the same data base is used, summation codes should yield essentially the same results irrespective of the different numerical methods used. The ENDF/B-IV data base was rapidly incorporated within other summation codes. After the release of the ENDF/B-IV fission product data file the majority of publications relating to the decay heat calculation originated in the USA and used this data base in 1970 s. Towards the end of 1978 results were available for most of ^{235}U and ^{239}Pu decay heat measurements[31-39] in progress. A summary of comparisons of these results with results of ENDF/B-IV calculations was given by England et al. (1978)[40]. Comparison results were encouraging, and confidence grew in summation method with the ENDF/B-IV data base. Efforts in the USA turned to the development of an updated fission product data library —— ENDF/B-V.

In 1977 the IAEA held it second Advisory Group Meeting on Fission Product Nuclear Data, at which Schenter et al.[7] reviewed the calculational and experimental progresses achieved in the determination of decay heat. It was concluded that the summation method for calculating fission product decay heat was both valid and rapidly improving in accuracy. Nevertheless, there remained discrepancies between calculations based on different data libraries, and between different experimental results which required resolving. The need for further improvements in data libraries was also emphasized.

There are many known, and unknown, short-lived fission products for which no detailed beta or gamma data are available. In these cases, theoretical estimates of mean beta and gamma energies are made with the aid of Q value for beta decay. The energy partition between the unknown beta, gamma and antineutrino has been estimated in a number of different ways. For example, Tobias (1973)[41] and Blachot and Fiche (1978)[42] divide the Q value into three equal quantities, Tasaka and Sasamoto (1974)[26] base their estimates on averaging of known decay energies with weights of activities of fission products at short cooling times, England and Schenter (1975)[28] and Tasaka (1979)[43] base their estimates on systematics, while Yoshida (1977)[44] has derived an energy partition using the gross theory of beta decay as developed by Takahashi and Yamada (1969)[45] and Koyama, Takahashi and Yamada (1970)[46].

Works in 1980 s

In 1980 s the gross theory of beta decay was extensively used in Japan[47,48] to estimate the decay data for short-lived fission products, and the agreement between calculated and measured decay heat was remarkably improved at short cooling times less than 10^3 s.

Yoshida and Nakasima (1981)[47] estimated unknow decay data, i.e. decay constant and mean beta (E_β) and gamma (E_γ) decay energies per disintegration, by the gross theory. They also replaced the experimental values of E_β and E_γ by the theoretical values calculated with the gross theory for 87 short-lived fission products with Q values larger than 5 MeV. This is because the theoretical values of E_β and E_γ for high Q value nuclides are more reliable than the experimental values when evaluated for the whole nuclides but not the individual. The energy levels at high energies are too dense and complicated for existing gamma-ray detectors to detect and resolve all transitions from them. The oversimplification of the high energy part leads to an underestimation or missing of the beta strength at high energies, and leads to an overestimation of

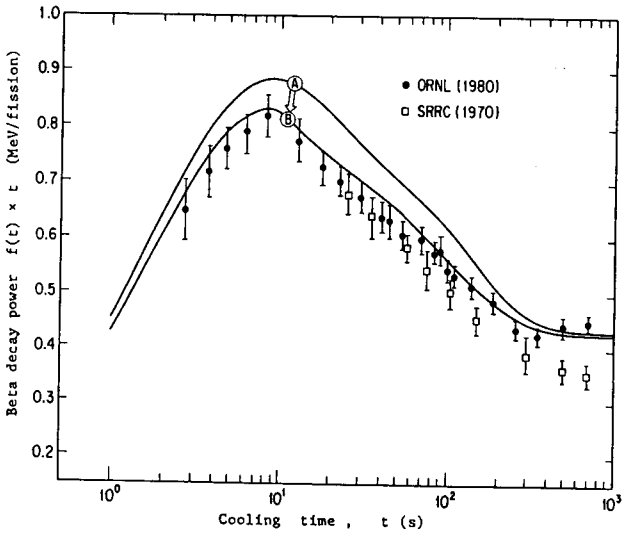

Fig. 1 Effect of using theoretical values of E_β and E_γ on the calculated results of ^{235}U beta decay heat. The solid curve with Ⓑ corresponds to calculation after the replacement of experimental E_β and E_γ values for 87 nuclides with theoretically estimated ones. The solid curve with Ⓐ corresponds to calculation before the replacement (from Tasaka et al./48/).

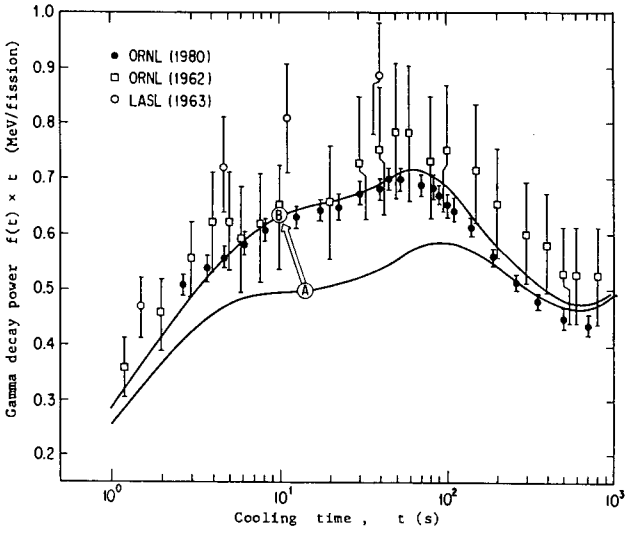

Fig. 2 Same as in Fig. 1, but for gamma decay heat.

E_β and underestimation of E_γ. Therefore, a full adoption of experiment-based values of E_β and E_γ results in an overestimation of the beta-ray component of decay heat and an underestimation of the gamma-ray component as clearly shown by solid line with Ⓐ in Figs. 1 and 2. This defect was resolved in the first version of the JNDC (Japanese Nuclear Data Committee) FP Nuclear Data Library by replacing the experimental values of E_β and E_γ by the theoretical values as mentioned above. The accuracy of the decay heat calculation was greatly improved not only for ^{235}U as shown by solid line with Ⓑ in Figs. 1 and 2 but also for other fissile nuclides.

The decay data of all experimentally identified fission product nuclides included in the JNDC FP Nuclear Data Library were reviewed in detail later on, since the missing of beta-transition to unobserved highly excited states in the daughter nucleus is considered to be probable in some cases even for nuclides with small Q_β. Thus the decay energies of 127 nuclides, except for ^{88}Rb and ^{143}La revised previously[49,50], were reevaluated[51,52]. The results of summation calculations based on the revised library (JNDC FP Nuclear Data Library Version 2)[52] became in much better agreement with experimentally measured decay heat curves than previous ones[53]. Especially, the discrepancy remained for cooling times longer than a few hundreds seconds was removed.

Tasaka et al. (1987)[53] calculated the decay heat of fission products for various fissile nuclides using the DCHAIN code[16,17] and the three different libraries; JNDC-V1, -V2 and ENDF/B-V. Calculated results were compared with measured results for fast neutron fission of ^{232}Th, ^{233}U, ^{235}U, ^{238}U and ^{239}Pu at the fast neutron source reactor YAYOI of University of Tokyo by Akiyama et al.[54-57] and also with measured ones at ORNL by Dickens et al.[33-36] for thermal neutron fission of ^{235}U, ^{239}Pu and ^{241}Pu.

Figures 3 through 5 show the comparisons of calculated and measured decay heats of beta, gamma, and beta plus gamma rays by fast-neutron fissions of ^{235}U. The abscissa is the time t after burst fission and the ordinate is the decay heat multiplied by the time t. As is seen in the figures the calculation by JNDC-V2 agrees satisfactorily with the measured data within ±4% for the total decay heats, ±5% for beta decay heats and ±5% for gamma decay heats. The measurement uncertainties (1σ) are estimated to be 5% for both beta and gamma decay heats. The calculated decay heats by the present JNDC-V2 file are in much better agreement with the measured ones than calculation by the ENDF/B-V especially at short cooling times less than 10^3 s. Although the discrepancies of the calculated results by ENDF/B-V from the measured ones are rather small for beta, and beta plus gamma decay heats, 8% overestimation and 8% underestimation respectively, considerable discrepancies of as much as 22% are seen in Fig. 4 for the gamma decay heats. The improvement in JNDC-V2 is also noticeable compared to the calculation by JNDC-V1 at longer cooling times than 300 s. The maximum discrepancies between the calculated decay heats by JNDC-V1 and the measured ones at cooling times greater than 300 s are 10%, 10% and 5%, respectively for beta, gamma and beta plus gamma decay heats.

For fast-neutron fissions of ^{239}Pu, ^{238}U, ^{233}U and ^{232}Th, the calculated decay heats by JNDC-V2 agree very well within ±5% with the

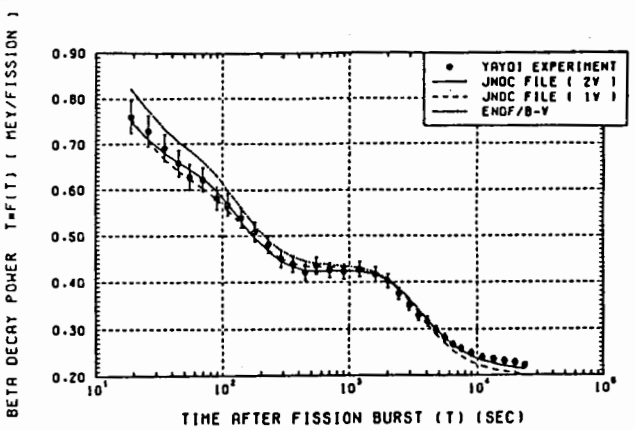

Fig. 3 Comparison of calculated beta decay heat to measured results at University of Tokyo for burst fission of ^{235}U by fast neutrons (from Tasaka et al./53/).

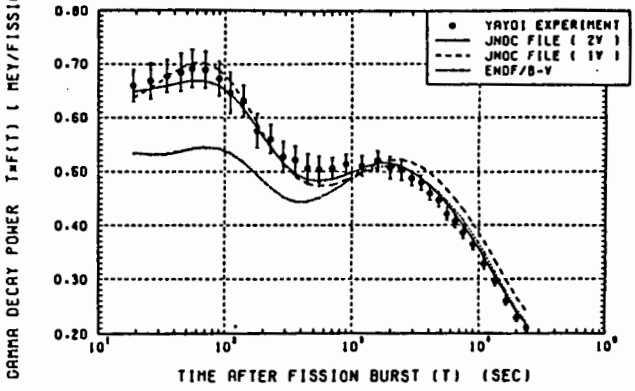

Fig. 4 Same as in Fig. 3, but for gamma decay heat.

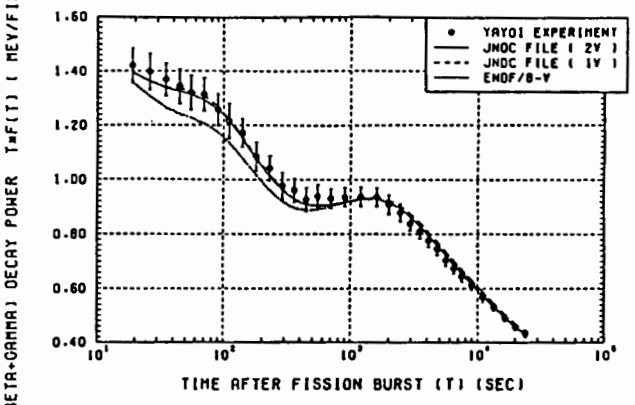

Fig. 5 Same as in Fig. 3, but for beta plus gamma decay heat.

measured ones with some exceptions. The calculation for ^{238}U underestimates the decay heat by 7% around cooling times of 10^3 s and the calculation for ^{232}Th underestimates by 12% and 15% at cooling times less than 10^2 s and greater than 10^4 s, respectively, due possibly to insufficient accuracies in the fission yield data for ^{232}Th. It was concluded at the 1987 Studsvik meeting[58], that the JNDC library is the only library to be able to give good agreement between summation calculations and integral measurements at short cooling times.

Klapdor (1985)[59] and Metzinger and Klapdor (1985)[60] estimated unknown decay data by

microscopic nuclear structure calculations for beta decays, and calculated the decay heat of fission products using the THOR-I code for short time irradiation. The calculated results for ^{235}U, ^{239}Pu, ^{241}Pu and ^{233}U are compared with measured results by Akiyama et al.[54-57] and Dickens et al.[33-36] as shown in Fig. 6 for example. The calculations reproduce the experiments reasonably well within the error bars of the experiments for fissions of ^{233}U and ^{235}U, whereas some underpredictions from measured results are seen for fissions of ^{239}Pu and ^{241}Pu around the cooling times of 10 s and 10^3 s with the maximum discrepancies of 10%. The results are not shown for ^{232}Th and ^{238}U.

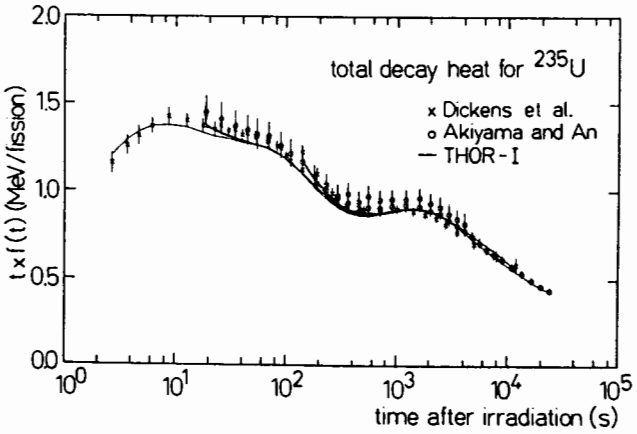

Fig. 6 Measured beta plus gamma decay heat for short-time irradiation of ^{235}U and corresponding calculations by THOR-I (from Metzinger and Klapdor/60/).

It is considered possible now to establish the decay heat standard based on the latest summation calculations with careful estimations of unknown or unreliable nuclear data of individual fission products. At the same time accurate measurements of unknown beta and gamma decay energies of short-lived fission products are recommended to improve further the reliability of the summation calculation and also to extend the application area of decay heat calculation. From this standpoint the measurement of beta decay energies of individual nuclides by Rudstam et al. (1978)[61] is important and noteworthy.

Gamma-Ray Spectrum

The importance and capability of the summation calculations of gamma-ray spectra were realized shortly after one of the first summation calculations of the decay heat following ^{235}U thermal fission. The gamma spectra are indispensable for shielding calculations. The first such gamma spectrum calculations would be those of Moteff (1953)[62], which used a data library of only 24 nuclides and yielded results for ^{235}U thermal fission at shutdown times in excess of 4 h.

The data library was extended with time to include decay data of more fission products and the capability of calculating the gamma spectrum was gradually improved in accuracy and also extended to shorter as well as longer cooling times. It was necessary to estimate the unknown decay data, especially gamma-ray spectrum data, of the individual fission product nuclide by theory or statistics[63-65] as well as to accumulate steadily the measured gamma-ray spectrum data of the individual nuclide.

Comparisons between ENDF/B-IV calculations and measurements of fission-product gamma spectra were made by England and Stamatelatos (1977)[66] and by Jurney et al. (1979)[67]. The decay heat measurements of Yarnell and Bendt[37,38] and Dickens et al.[33-36] included also the measurement of gamma spectra. The ENDF/B-IV decay data library contained beta and gamma spectra data only for 180 of the nuclides in the library. Therefore, it was assumed that the gamma spectral shapes derived from these data adequately represented those for the complete set of fission products, even at short cooling times when the 180 nuclides contribute only a fraction of the total gamma energy. The comparisons proved much better than had been originally anticipated, however, some short comings were found for short cooling times especially in the gamma-ray energy range of 1.5 - 3.0 MeV.

It was necessary to wait until 1986 to have reasonable agreement between calculated gamma spectra and measured data for a wide range of cooling time including a very short cooling time. Yoshida and Katakura (1986)[64] and Katakura and Yoshida (1988)[65] estimated the gamma-ray spectra of the unstable nuclides without experimental spectrum data by the calculation based on the beta strength function derived from the gross theory of beta decay and on the cascade gamma-ray transition model[64]. In the scheme of the gross theory, the beta strength function varies slowly with mass number A and is quite sensitive to evenness and oddness of the proton number Z and the neutron number N through the pairing correlation. Therefore, the gamma-ray spectrum estimations were performed for typical 32 nuclides categolized by mass number (light, e.g., A=95 or 96 and heavy, e.g., A=139 or 140), by odd-even type of the proton and the neutron numbers (odd-odd, odd-even, even-odd and even-even) and by Q_β value (5, 7, 9 and 11 MeV).

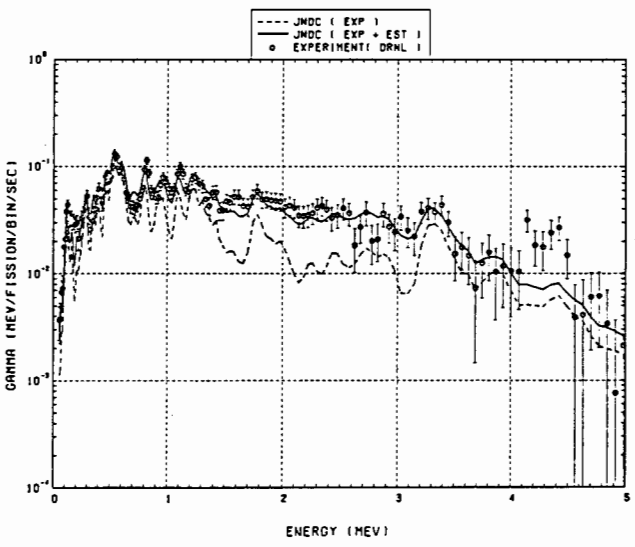

Fig. 7 Gamma-ray energy spectrum at 2.7 s after instantaneous fission of ^{235}U by thermal neutron. Dotted line is the calculation where only experimental spectra of individual nuclides are included. Solid line indicates the calculation compensated by the estimated spectra of nuclides without experimental spectral data. Open circules are the measurements at ORNL (from Katakura and Yoshida/65/).

Figure 7 exemplifies the calculated gamma-ray spectrum from aggregate fission product nuclides compensated by the estimated spectra comparing with the measured spectrum at 2.7 s after the thermal neutron fissions of ^{235}U. The calculations by using only the experimental spectrum data are also shown by dotted lines. As seen in the figure, the effect of the compensation by the estimated spectra are apparent. Agreement becomes more satisfactory at longer cooling times.

Effect of Neutron Captures in Fission Products

The effect of neutron captures in fission products on the decay heat had been noticed since 1950 s[22], shortly after the calculation of the decay heat was initiated by the summation method. The effect can be neglected for short time irratiation in low level neutron fluxes, however, it is necessary to consider the effect for typical reactor operating conditions.

One of the first summation studies to include the effect of neutron capture, and investigate its influence on the decay heat, was that of Perkins and King (1958)[23], in which neutron capture only in ^{135}Xe was considered. The effect was found negligible in the cases studied by Perkins and King. As a result, it was some years before neutron capture effects were studied again in detail, when England (1969)[68] made a detailed study of decay heat, including the effect of neutron capture, using the code CINDER[69] and its 350-nuclide data library. The neutron capture effects were found substantial under certain conditions and the results implied that the ^{235}U decay heat curve derived by Shure (1961)[2] for an infinite irradiation could underestimate the decay heat in realistic reactor operation conditions. These findings, although later found to be in error, had an important effect in stimulating the activity in further studies of decay heat.

One of the most detailed studies of neutron capture effects on decay heat was made by Tasaka (1977)[70]. Calculations were made both with and without neutron capture reactions for the irradiation of fuel from typical LWR, GCR and LMFBR. As in the other studies[3,24], the effect on decay heat was found to be negligible for times less than 10^4 s. At the longer cooling times, when the effect becomes significant, it is due to a relatively small number of nuclides—particularly shielded nuclides such as ^{134}Cs, ^{136}Cs, ^{148m}Pm, ^{148}Pm and ^{154}Eu. He also investigated the effect of using different data libraries, flux levels and irradiation times. The major differences found with the various data libraries were due almost entirely to the differences in the value of the ^{133}Cs neutron-capture cross section. It was concluded that generally the capture effect increases with an increase in either flux level or irradiation time. At the longer cooling times the effect was found to be approximately proportional to the product of neutron flux and irradiation time.

Although the influence of neutron capture on fission product decay heat has been found to be small for short cooling times, some efforts[71,72] have been made to model these effects for times less than 10^4 s. The model recommended by Spinrad and Tripathi (1978)[72] is

$$C = (3.24 \times 10^{-4} + 5.23 \times 10^{-8} \times t)T^{0.4} \times (\text{FIFA}) \quad (31)$$

where C is the percentage increase in decay heat, T is the irradiation time in s, t is the cooling time in s, and (FIFA) is the number of fissions per initial fissile atom.

For longer cooling times, it is possible to calculate the effect of neutron capture by considering only a few of the nuclides. Such an approach has been adopted by La Bauve et al.[73,74], who considered only the more important capture chains to supplement the decay heat results derived from their exponential fits.

A simplified method was proposed by Tasaka and Iijima (1986)[75] for the calculation of the effects of neutron capture transformations of fission products on the decay heat. It was proposed to calculate the neutron capture transformation effects considering the production of only 7 nuclides ^{103}Ru, ^{134}Cs, ^{136}Cs, ^{148m}Pm, ^{148}Pm, ^{154}Eu and ^{156}Eu by the neutron capture reaction of the direct mother nuclide alone giving a cumulative fission yield for the mother nuclide. The method was assessed by comparing the calculation results with the rigorous calculation results for the fission of ^{235}U ^{238}U and ^{239}Pu irradiated between 1 and 5 y in a light water reactor with thermal-neutron flux between 3×10^{13} and 6×10^{13} n/cm^2 s and for the fission of ^{239}Pu, ^{241}Pu and ^{238}U irradiated between 1 and 5 y in a fast breeder reactor with total neutron flux between 3×10^{15} and 6×10^{15} n/cm^2 s. It has been clarified that the method with the simple correction factors can calculate the neutron capture transformation effects within the accuracy of $\pm 1\%$ of the decay heat for the irradiation of 1 - 5 y and cooling time less than 10^9 s irrespective of fission type and neutron flux (see Fig. 8 as an example).

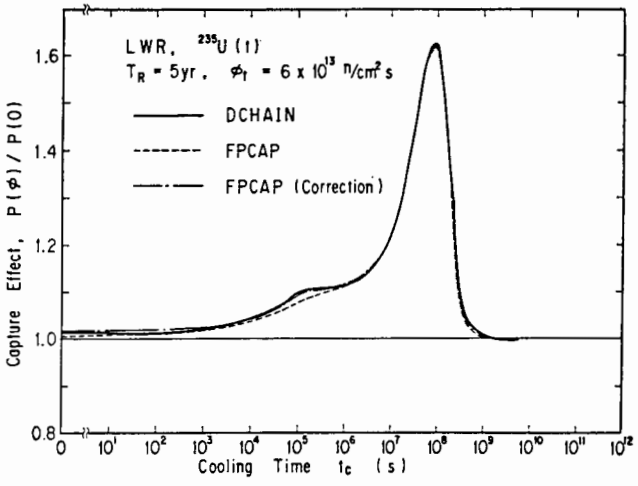

Fig. 8 Neutron capture transformation effects of FPs on decay heat of FPs. The solid line shows rigorous results by DCHAIN, dashed line approx. results by FPCAP, and dash-dot line corrected approx. results (from Tasaka and Iijima/75/).

An integral measurement of the neutron capture effect or the decay heat itself is necessary with relatively long irradiation at high neutron flux to assess the accuracy of the neutron-capture-effect calculation method, because the integral measurement of decay heat was conducted with relatively short irradiation at low neutron flux and included little neutron capture contribution up to now.

Unceratinties in Decay Heat Summation Calculations

The reliability of the summation method of decay heat calculation can be evaluated qualitatively by comparing calculated results with corresponding experimental results. It is also possible to evaluate directly the unceratinty in decay heat predictions due to the uncertainties in data input to the summation code. The benefits of such analyses are three-fold. Firstly, the uncertainty evaluation makes it possible to determine the conservative decay heat curve for a LWR LOCA analysis and other applications. Secondly, the sensitivity studies are capable of identifying those areas of basic data which have the greatest impact on the accuracy of summation calculations and which should therefore have the highest priority in future evaluation work. Thirdly, an independent evaluation of the uncertainties in summation calculations permits one to assess the available experimental results, particularly from the point of view of systematic biases to which the measurements are susceptible.

A number of studies have been made of the sensitivity of summation calculations to uncertainties in fission product yields, half-lives and decay energies between 1975 and 1978[47,76-83] in accordance with the recommendation by Lott (1973)[4].

Schmittroth and Schenter (1977)[77] evaluated decay heat uncertainties from the uncertainties of decay energies, fission yields and half-lives of individual fission products for ^{235}U thermal fission. The results for the burst fissions are shown in Fig. 9. It is seen in the figure that the decay heat uncertainties at short cooling times < 10^2 s are primarily due to uncertainties in decay energies and that the uncertainty decreases to a minimum value in the time range 10^3 - 10^6 s, before increasing again at the longer

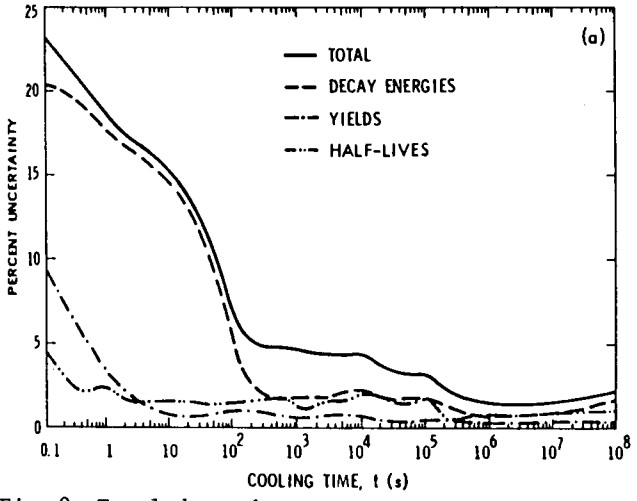

Fig. 9 Total decay heat uncertainties for thermal fission of ^{235}U burst exposure (from Schmittroth and Schenter/77/).

cooling times. This minimum arises as a consequence of the important decay heat nuclides being both well known and numerous.

Tobias (1980)[10] compared the total uncertainties estimated in summation results for different thermal fission processes for irradiation times of 10^7 - 10^9 s. The results by Devillers (1977)[82] are much smaller than the other results at short cooling times less than 10^3 s perhaps due to the fact that Devillers did not

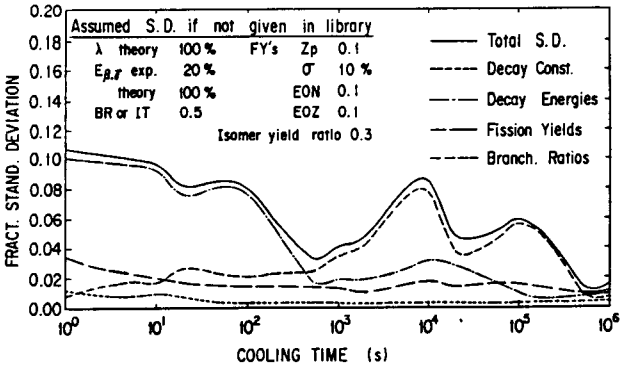

Fig. 10 Fractional standard deviation of decay heat from one fission of ^{235}U (from James/84/).

treat the correlated uncertainties in decay energies exactly. At cooling times longer than 10^3 s the contribution of decay energy uncertainties to the total uncertainties becomes smaller and the differences among different fission types and evaluations become smaller.

James (1987)[84] evaluated resently the uncertainties of decay heat calculations by the summation method as shown in Fig. 10 for burst fissions of ^{235}U by thermal neutrons. The uncertainties at short cooling times less than 10^3 s become smaller than the results by Schmittroth and Schenter[77] in 1977 due to improvement of accuracies of decay energies of short-lived fission products. The figure also illustrates the considerable contribution of branching fraction uncertainties to the total decay heat uncertainties at cooling times longer than 10^3 s resulting in the maximum uncertainty of as much as 8%.

Conclusions

Following conclusions were obtained in the present review of decay heat calculations of fission products:

(1) The calculated decay heat by the summation method using the JNDC-V2 library, agreed within the accuracy of ±5% with the measured data at the University of Tokyo for the fast-neutron fission of ^{233}U, ^{235}U, ^{238}U, ^{239}Pu and the data at ORNL for the thermal-neutron fission of ^{239}Pu and ^{241}Pu. In the JNDC-V2 library unknown or unreliable decay data, especially decay energies, of short-lived nuclides were estimated by the gross theory of beta decay. Discrepancies of greater than 5% still remains for ^{232}Th fast fission due possibly to insufficient accuracis for yields data for ^{232}Th fast fission.

(2) The calculated gamma-ray spectrum by the summation method using the JNDC-V2 library agreed reasonably well with measured data at the University of Tokyo and ORNL. In the JNDC-V2 library, unknown gamma-ray spectrum data of individual fission product nuclide were estimated by the cascade gamma-ray transition model with the beta strength function derived from the gross theory of beta decay.

(3) The neutron capture effect of fission products on the decay heat is small for the cooling times < 10^4 s, however, the effect can become considerable at longer cooling times. Therefore, the integral measurement of the capture effect or of the decay heat including a considerable capture effect is recommended for assessing the accuracy of the capture effect calculation.

(4) A continuous effort is necessary to improve the accuracy and to extend the range of data base by the experiment or the theory, in order to resolve existing deficiencies and to extend the application. Also integral measurements of decay heat are necessary for assessing the accuracy of calculation in extended application areas.
(5) The decay heat standard is under preparation in Japan, based on the summation calculations using the JNDC-V2 nuclear data library for fission products. The standard will include expressions for gamma spectrum and neutron capture effect as well as beta, gamma and beta plus gamma decay heat with uncertainties for a variety of fissile nuclides.

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