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New Method for Calculating Aggregate Fission Product Decay Heat with Full Use of Macroscopic-Measurement Data

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We propose a new "hybrid" method for calculating the aggregate decay heat from fission product nuclides after a fission burst. The decay heat from a given fissioning system is expressed as a linear combination of macroscopic-measurement data for other fissioning systems with a small residual term. This method is based on the linearity of the decay heat to the fission yield. The coefficients in the linear combination are obtained from fitting the fission yield of the given fissioning system with a linear combination of fission yields of other fissioning systems. To demonstrate usefulness of this method, it is applied to examining the consistency among measured decay heat powers of five fast and three thermal neutron induced fissions. The hybrid-method calculations agree well with the measurements and usual summation calculations at cooling times before 4,000 s, except for a γ component measurement of the ^{235}U thermal fission at about 2,000 s. These results indicate the consistency and reliability of the decay heat evaluation for these systems with the above exception. Furthermore, they also imply usefulness of the present method in predicting the decay heat of other fissioning systems, for which no measurements have been performed so far.

KEYWORDS: decay heat, decay heat measurements, decay heat removal, summation method, fission products, fission yields, decay data, reliability

I. Introduction

The aggregate fission product (FP) decay heat is one of the most important nuclear properties required in all systems that utilize the nuclear fissions. The decay heat should be evaluated not only to operate nuclear reactors but also to reprocess, transport and dispose radioactive materials produced in the reactors. It should be noted that the evaluations should not be limited to fissile nuclides in the initial inventory since other fissile nuclides are produced through transmutation processes in nuclear reactors.

In this paper, we focus on the aggregate FP decay heat power after a fission burst of a fissile nuclide. The neutron absorptions in FP's, which are dependent on irradiation history, never occur in this case. In the following, the FP decay heat after a fission burst is referred to as the decay heat for simplicity.

The evaluations of the decay heat have been performed in two methods. One is the macroscopic measurement in which we directly measure the aggregate decay heat from a sample containing a fissile nuclide after appropriate irradiation. The other is the (microscopic) summation method, in which we calculate the decay and buildup of about thousand FP nuclides from their fission yields and decay data. To date, several measurements have been performed in limited cooling time ranges for major fissile nuclides while input nuclear databases have been compiled for summation calculations. Now, various measurements and calculations come to give more or less similar results.

However, the reliability of the present decay heat evaluation in the two methods could be worse than we have expected. In the summation calculations, most of the FP's are so short-lived that their fission yields and decay data in the input nuclear database are not known well. Actually, many of the input data have to be estimated theoretically or from systematics so that their uncertainties are not known. Hence, the success of summation calculations might be a consequence of cancellation effects among wrong input FP data thanks to some gross nuclear properties. Moreover, as we will see later, the measurements (and/or calculations) for some cases still show distinct deviations beyond their uncertainties. Therefore, it is essential, even in the latest decay heat evaluation, to find out consistent values from various macroscopic-measurement data and summation calculations obtained from many uncertain FP data.

In this paper, we propose a new method for calculating the aggregate decay heat power, which can be used to examine consistency among macroscopic-measurement data. The present method can be used to choose a set of reliable measurement data, to find out potential inconsistency among them, and/or to obtain some information about fission yields or decay data of FP nuclides for summation calculations.

The present method is based on the fact that the decay heat after a fission burst is a linear function of the fission yields of the FP nuclides. We utilize the fact that the fission yields are more or less similar among different fissioning systems.[1,2] In this method, the decay heat power of a fissioning system is written as a linear combination of those of other fissioning

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systems with a small residual term. The coefficients of the combination are calculated from the fission yields. Then, the residual term is obtained from summation calculations using both the fission yields and decay data of FP's.

To demonstrate usefulness of the hybrid method, it is applied to examining the consistency among macroscopic-measurement data of contemporary use. They are the decay heat powers for five fast and three thermal fissioning systems, that were measured at YAYOI reactor of the University of Tokyo[3–6] and at Oak Ridge National Laboratory (ORNL),[7,8] respectively. The FP fission yields and decay data needed for this analysis are taken from ENDF/B-VI.[2,9]

This paper is arranged as follows. In Chap. II, the summation method is briefly reviewed in the matrix representation. In Chap. III, our new method, referred to as hybrid method, is developed using formula in the matrix representation. In Chap. IV, the consistency analysis in the hybrid method is demonstrated among the macroscopic-measurement data. Chapter V is devoted to discussions. Finally, the conclusion of this paper is given in Chap. VI.

II. Summation Method in the Matrix Representation

In this chapter, we briefly review the summation calculation of the decay heat power after a fission burst in the matrix representation. The decay heat power $P(t)$ at a cooling time t is given as the sum of energy releases per unit time from individual FP nuclides,

$$P(t) = \sum_{i=1}^{M} E_i \lambda_i N_i(t), \tag{1}$$

where λ_i and E_i are the decay constant and average decay energy per decay of nuclide i, respectively. The number of the FP nuclides is denoted by M. The atom number of nuclide i at cooling time t, $N_i(t)$, is a solution of coupled differential equations that describe decays and buildups of the M FP nuclides;

$$\frac{d}{dt} N_i(t) = -\lambda_i N_i(t) + \sum_{j \neq i} b_{j\to i} \lambda_j N_j(t), \tag{2}$$

with

$$N_i(0) = y_i. \tag{3}$$

Here, y_i and $b_{j\to i} (\geq 0)$ are the independent fission yield of nuclide i, and the branching ratio to nuclide i per decay of nuclide j, respectively.

It is noted that Eq. (2) is a linear differential equation. Therefore, we can solve them formally using vectors of $N_i(t)$'s and y_i's, and matrices of the decay constants λ_i's and branching ratios $b_{j\to i}$'s. The vectors of the atom numbers and the fission yields are given by

$$\boldsymbol{N}(t) = \begin{pmatrix} N_1(t) \\ N_2(t) \\ \vdots \\ N_M(t) \end{pmatrix} \tag{4}$$

and

$$\boldsymbol{y} = \begin{pmatrix} y_1(t) \\ y_2(t) \\ \vdots \\ y_M(t) \end{pmatrix}, \tag{5}$$

respectively. The matrices of the decay constants and branching ratios are defined as

$$\boldsymbol{\Lambda}_0 = \begin{pmatrix} \lambda_1 & 0 & \cdots & 0 \\ 0 & \lambda_2 & \cdots & 0 \\ \vdots & \vdots & \ddots & \vdots \\ 0 & 0 & \cdots & \lambda_M \end{pmatrix} \tag{6}$$

and

$$\boldsymbol{B} = \begin{pmatrix} 0 & b_{2\to 1} & \cdots & b_{M\to 1} \\ b_{1\to 2} & 0 & \cdots & b_{M\to 2} \\ \vdots & \vdots & \ddots & \vdots \\ b_{1\to M} & b_{2\to M} & \cdots & 0 \end{pmatrix}, \tag{7}$$

respectively. Then, Eq. (2) can be written simply as

$$\frac{d}{dt}\boldsymbol{N}(t) = (\boldsymbol{B} - \boldsymbol{I})\boldsymbol{\Lambda}_0 \boldsymbol{N}(t), \tag{8}$$

where the symbol $\boldsymbol{I}$ denotes the unit matrix of order M. The initial condition (3) is now given by

$$\boldsymbol{N}(0) = \boldsymbol{y}. \tag{9}$$

The formal solution of Eq. (8) with initial condition (9) is simply written as

$$\boldsymbol{N}(t) = \exp((\boldsymbol{B} - \boldsymbol{I})\boldsymbol{\Lambda}_0 t)\boldsymbol{y}. \tag{10}$$

Here, the exponential function of a matrix $\boldsymbol{A}$ is defined as

$$\exp(\boldsymbol{A}) = \boldsymbol{I} + \boldsymbol{A} + \frac{1}{2!}\boldsymbol{A}^2 + \frac{1}{3!}\boldsymbol{A}^3 + \frac{1}{4!}\boldsymbol{A}^4 + \cdots. \tag{11}$$

We note that the solution (10) is a linear function of the fission yield (vector) $\boldsymbol{y}$. Furthermore, the decay heat power $P(t)$ is also a linear function of the fission yield $\boldsymbol{y}$. In fact, Eq. (1) can be written, using Eq. (10), as

$$P(t) = (E_1 E_2 \cdots E_M)\boldsymbol{\Lambda}_0 \exp((\boldsymbol{B} - \boldsymbol{I})\boldsymbol{\Lambda}_0 t)\boldsymbol{y}. \tag{12}$$

From the formal solutions (10) and (12), we see clearly that $\boldsymbol{N}(t)$ and $P(t)$ are obtained as products of the fission yield ($\boldsymbol{y}$) and linear operators composed of decay data ($\boldsymbol{\Lambda}_0$, $\boldsymbol{B}$ and average decay energies). It is only the fission yield $\boldsymbol{y}$ that distinguishes fissioning systems. It is also noted that we use the same decay operators independently of fissioning systems.

III. Hybrid Method for Calculating Decay Heat Power

The hybrid method makes full use of the fact that the decay heat is a linear function of the fission yield $\boldsymbol{y}$. We note that the fission yield vector $\boldsymbol{y}$ for a major fissile nuclide is a gradual and relatively regular function of the atomic mass and proton number in low energy neutron fissions (below a few MeV). Hence, the dominant part of the fission yield $\boldsymbol{y}$ is common among major fissioning systems and thus can be approximated well by a linear combination of fission yield vectors of other fissioning systems. Consequently, thanks to the linearity, we can write the decay heat power of a fissioning system as a linear combination of others' with a small residual term.

Initially, we approximate a fission yield vector $\boldsymbol{y}$ of a fissioning system by a linear combination of fission yield vectors of other fissioning systems. For example, let us write $\boldsymbol{y}$ with a linear combination of fission yield vectors of four other fissioning systems, $\boldsymbol{y}_1 - \boldsymbol{y}_4$;

$$\boldsymbol{y} = a_1\boldsymbol{y}_1 + a_2\boldsymbol{y}_2 + a_3\boldsymbol{y}_3 + a_4\boldsymbol{y}_4 + \boldsymbol{y}_R. \tag{13}$$

Here, $\boldsymbol{y}_R$ denotes the residual term that cannot be represented by the four fission yield vectors. Values of the four coefficients, $a_1 - a_4$, are determined to minimize the absolute value of the residual term, $|\boldsymbol{y}_R|$. Then, the coefficients are written explicitly as

$$\begin{pmatrix} a_1 \\ a_2 \\ a_3 \\ a_4 \end{pmatrix} = \begin{pmatrix} \boldsymbol{y}_1\cdot\boldsymbol{y}_1 & \boldsymbol{y}_1\cdot\boldsymbol{y}_2 & \boldsymbol{y}_1\cdot\boldsymbol{y}_3 & \boldsymbol{y}_1\cdot\boldsymbol{y}_4 \\ \boldsymbol{y}_2\cdot\boldsymbol{y}_1 & \boldsymbol{y}_2\cdot\boldsymbol{y}_2 & \boldsymbol{y}_2\cdot\boldsymbol{y}_3 & \boldsymbol{y}_2\cdot\boldsymbol{y}_4 \\ \boldsymbol{y}_3\cdot\boldsymbol{y}_1 & \boldsymbol{y}_3\cdot\boldsymbol{y}_2 & \boldsymbol{y}_3\cdot\boldsymbol{y}_3 & \boldsymbol{y}_3\cdot\boldsymbol{y}_4 \\ \boldsymbol{y}_4\cdot\boldsymbol{y}_1 & \boldsymbol{y}_4\cdot\boldsymbol{y}_2 & \boldsymbol{y}_4\cdot\boldsymbol{y}_3 & \boldsymbol{y}_4\cdot\boldsymbol{y}_4 \end{pmatrix}^{-1} \begin{pmatrix} \boldsymbol{y}_1\cdot\boldsymbol{y} \\ \boldsymbol{y}_2\cdot\boldsymbol{y} \\ \boldsymbol{y}_3\cdot\boldsymbol{y} \\ \boldsymbol{y}_4\cdot\boldsymbol{y} \end{pmatrix}. \tag{14}$$

The derivation of Eq. (14) is described in **Appendix**. The coefficient values are determined from the inner products of the vectors. Hence these values reflect differences in gross properties of the fission yield vectors.

Then, from the linearity to the fission yield $\boldsymbol{y}$, the decay heat power $P(t)$ can also be given, using the same coefficients, by a linear combination of decay heat powers of the four fissioning systems, $P_1(t) - P_4(t)$;

$$P(t) = a_1P_1(t) + a_2P_2(t) + a_3P_3(t) + a_4P_4(t) + P_R(t). \tag{15}$$

Here, the residual term $P_R(t)$ can be calculated from the residual yield $\boldsymbol{y}_R$ in the summation method;

$$P_R(t) = (E_1E_2\ldots E_M)\boldsymbol{\Lambda}_0\exp((\boldsymbol{B}-\boldsymbol{I})\boldsymbol{\Lambda}_0t)\boldsymbol{y}_R. \tag{16}$$

The $P_R(t)$ value is expected to be small because $|\boldsymbol{y}_R|$ is minimized. Practically, we do not have to use Eq. (16) to calculate the residual term $P_R(t)$. Instead, once we calculate $P(t)$ and $P_1(t) - P_4(t)$ in the summation method, $P_R(t)$ is given by

$$P_R(t) = a_1P_1^c(t) + a_2P_2^c(t) + a_3P_3^c(t) + a_4P_4^c(t) - P^c(t). \tag{17}$$

The superscript "c" denotes the summation calculation.

Equations (14), (15) and (17) constitute a set of working formula to calculate the decay heat power $P(t)$ in the present method. From the fission yields of the five systems, we can calculate the coefficient values using Eq. (14). Then, the decay heat power $P(t)$ is obtained from Eq. (15) by putting the measured decay heat powers of the other four systems into $P_1(t) - P_4(t)$. The value of the correction term, $P_R(t)$, can be calculated from Eq. (17) using both the fission yields and decay data. In the present method, we do not need the $\boldsymbol{y}_R$ value to calculate $P(t)$. If necessary, $\boldsymbol{y}_R$ can be calculated from Eq. (13).

The present method is hybrid between the macroscopic measurement and the summation calculation. The measured decay heat powers implicitly contain the true values of fission yields and decay data while the summation calculation of $P(t)$ requires evaluated fission yields and decay data for all FP nuclides. In the hybrid method, a dominant part of $P(t)$ is calculated from the measured decay heat powers of other fissioning systems, $P_1(t) - P_4(t)$. Only its small fraction, P_R, is calculated in the summation method. It is remarked that the decay data is minimally used only in the small residual term, P_R. This means that the hybrid method makes full use of the true decay data values that are implicitly contained in the measured decay heat powers. To make this clear, **Table 1** summarizes the relevant FP nuclear data to the decay heat evaluations in the three methods.

IV. Applications

The hybrid method is used to examine consistency among measured decay heat powers of fast and thermal neutron fissions. The measured powers in this chapter are given as the values for a fission burst so that the present method is directly applicable. **Figure 1** shows the outline of the consistency analysis schematically. A measurement data of a fissioning system is compared with a hybrid calculation (*i.e.*, calculation in the hybrid method) using data of the other fissioning systems. Furthermore, the hybrid calculations are also compared with summation calculations to see whether the overall consistency is satisfied among the values obtained in the three method. In the following, all values of the fission yields and decay data are taken from ENDF/B-VI.[2,9)]

Table 1 The fission product properties relevant to the decay heat evaluations in the three method

In the hybrid method, "evaluated" decay data are used only minimally so that the word "evaluated" is typed in the lower case.

	Summation method	Measurement	Hybrid method
Fission yields	Evaluated	True	Evaluated
Decay constants	Evaluated	True	True+evaluated
Branching ratios	Evaluated	True	True+evaluated
Average decay energies	Evaluated	True	True+evaluated

1. Consistency among YAYOI Measurements for Fast Neutron Fissions

The decay heat powers after fast neutron induced fissions of ^{232}Th, ^{233}U, ^{235}U, ^{238}U and ^{239}Pu were measured by Akiyama *et al*. at YAYOI reactor of the University of Tokyo.[3–6] Using the results of these measurements, we calculate a decay heat power of one of these fissiles from the rest in the hybrid method. We denote the measured (calculated) decay heat powers of ^{232}Th, ^{233}U, ^{235}U, ^{238}U and ^{239}Pu as P_1^m, P_2^m, P_3^m, P_4^m and P_5^m (P_1^c, P_2^c, P_3^c, P_4^c and P_5^c), respectively. Then, the relations among the decay heat powers (Eq. (15)) are written as

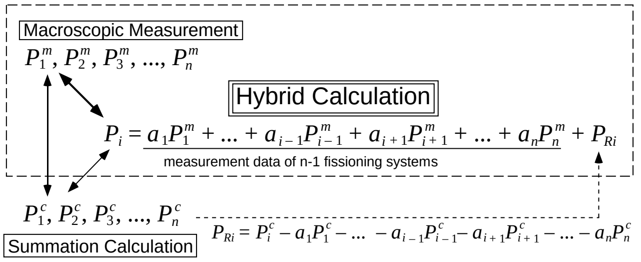

Fig. 1 The consistency analysis of macroscopic-measurement data of *n* fissioning systems

The symbol P_i^m denotes a measured decay heat power of the i-th fissioning system while P_i^c is the calculated one in the summation method.

$$P(^{232}\mathrm{Th}) = a_{12}P_2^m + a_{13}P_3^m + a_{14}P_4^m + a_{15}P_5^m + P_R(^{232}\mathrm{Th}), \tag{18}$$

$$P(^{233}\mathrm{U}) = a_{21}P_1^m + a_{23}P_3^m + a_{24}P_4^m + a_{25}P_5^m + P_R(^{233}\mathrm{U}), \tag{19}$$

$$P(^{235}\mathrm{U}) = a_{31}P_1^m + a_{32}P_2^m + a_{34}P_4^m + a_{35}P_5^m + P_R(^{235}\mathrm{U}), \tag{20}$$

$$P(^{238}\mathrm{U}) = a_{41}P_1^m + a_{42}P_2^m + a_{43}P_3^m + a_{45}P_5^m + P_R(^{238}\mathrm{U}), \tag{21}$$

$$P(^{239}\mathrm{Pu}) = a_{51}P_1^m + a_{52}P_2^m + a_{53}P_3^m + a_{54}P_4^m + P_R(^{239}\mathrm{Ph}). \tag{22}$$

In these equations, the cooling time t is suppressed for simplicity. The residual terms are obtained from summation calculations;

$$P_R(^{232}\mathrm{Th}) = - a_{12}P_2^c - a_{13}P_3^c - a_{14}P_4^c - a_{15}P_5^c + P_1^c, \tag{23}$$

$$P_R(^{233}\mathrm{U}) = -a_{21}P_1^c - a_{23}P_3^c - a_{24}P_4^c - a_{25}P_5^c + P_2^c, \tag{24}$$

$$P_R(^{235}\mathrm{U}) = -a_{31}P_1^c - a_{32}P_2^c - a_{34}P_4^c - a_{35}P_5^c + P_3^c, \tag{25}$$

$$P_R(^{238}\mathrm{U}) = -a_{41}P_1^c - a_{42}P_2^c - a_{43}P_3^c - a_{45}P_5^c + P_4^c, \tag{26}$$

$$P_R(^{239}\mathrm{Pu}) = -a_{51}P_1^c - a_{52}P_2^c - a_{53}P_3^c - a_{54}P_4^c + P_5^c. \tag{27}$$

Values of coefficients, a_{ij}, are calculated from the independent fission yields of these five systems using Eq. (14). **Table 2** lists the coefficient values. These values are not always positive. As a result, there are appreciable cancellations among measured decay heat powers, $P_1^m - P_5^m$, in Eqs. (18), (19), (21) and (22).

The decay heat powers calculated from Eqs. (18)–(22) are shown in **Fig. 2** for their β and γ components, separately. The residual term P_R is shown as the difference between the solid and dotted lines. From this figure, we see that the hybrid calculations agree with the macroscopic measurements reasonably within the experimental uncertainties except for cooling times longer than 4,000 s. The values of P_R/P are small, typically 10–20%, so that the above results are not very sensitive to uncertainties of the P_R values. Therefore we conclude that the measured decay heat powers at YAYOI are consistent with each other before 4,000 s.

On the contrary, after 4,000 s, there are systematic deviations from the measured values for all fissioning systems except for γ components of ^{232}Th and ^{239}Pu. This may imply that some of the measured values are inconsistent although the reason is not known at present.

As for the decay heat before 4,000 s, the results of the three methods agree reasonably with each other in Fig. 2. Therefore, we see that the decay heat powers in this cooling time range are established both experimentally and theoretically almost within the experimental uncertainties.

2. Consistency with ORNL Measurements for Thermal Neutron Fissions

Now we examine whether the ORNL measurements for the thermal fissions[7,8] are consistent with the YAYOI measurements for the fast fissions.

There has been no direct way to analyze the difference of decay heat powers between the thermal and fast fissioning systems. Consequently, it has often been assumed naively that the difference due to the neutron energy is small. In fact, the results of the YAYOI (ORNL) measurements have been used to discuss the decay heat powers of the thermal (fast) neutron fissions.

The hybrid method enables us to compare directly decay heat powers of fissioning systems with different neutron energies. Now, from the YAYOI values for the five fast systems, we calculate decay heat powers of the thermal neutron fissions of ^{235}U, ^{239}Pu and ^{241}Pu, which were measured at ORNL;

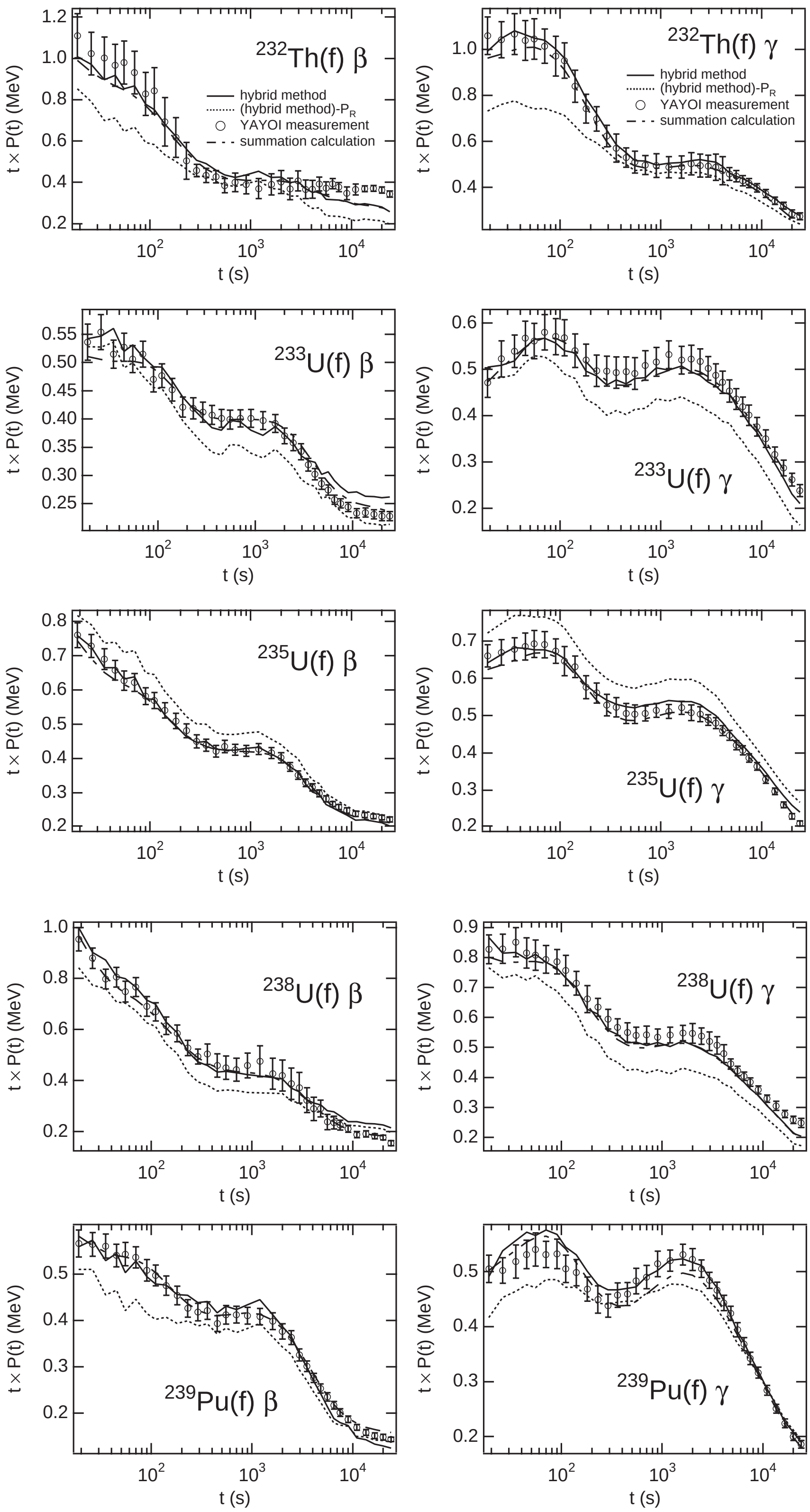

Fig. 2 Decay heat powers from pulse fast fissions of ^{232}Th, ^{233}U, ^{235}U, ^{238}U and ^{239}Pu

The solid lines show the $P(t)$ values obtained from Eqs. (18)–(22) while the dotted lines show the $P(t) - P_R(t)$ values. Also shown for comparison are the results of YAYOI measurements (open circles with error bars) and summation calculations with ENDF/B-VI (dash-dotted lines).

Table 2 Values of the coefficients a_{ij} to give relations among five fast fissioning systems of YAYOI measurement

i (fissile)	j				
	1 (^{232}Th)	2 (^{233}U)	3 (^{235}U)	4 (^{238}U)	5 (^{239}Pu)
1 (^{232}Th)	0	0.4196	0.3591	0.7948	−0.7115
2 (^{233}U)	0.1515	0	0.8166	−0.5385	0.4458
3 (^{235}U)	0.0805	0.5069	0	0.3888	0.1514
4 (^{238}U)	0.3567	−0.6692	0.7783	0	0.3761
5 (^{239}Pu)	−0.3063	0.5316	0.2909	0.3608	0

$$P(^{235}\mathrm{U}(t)) = b_{11}P_1^m + b_{12}P_2^m + b_{13}P_3^m + b_{14}P_4^m + b_{15}P_5^m + P_R(^{235}\mathrm{U(t)}), \quad (28)$$

$$P(^{239}\mathrm{Pu(t)}) = b_{21}P_1^m + b_{22}P_2^m + b_{23}P_3^m + b_{24}P_4^m + b_{25}P_5^m + P_R(^{239}\mathrm{Pu(t)}), \quad (29)$$

$$P(^{241}\mathrm{Pu(t)}) = b_{31}P_1^m + b_{32}P_2^m + b_{33}P_3^m + b_{34}P_4^m + b_{35}P_5^m + P_R(^{241}\mathrm{Pu(t)}). \quad (30)$$

Here, the symbol "t" denotes the thermal neutron induced fission. The residual terms are given by

$$P_R(^{235}\mathrm{U(t)}) = P^c(^{235}\mathrm{U(t)}) - b_{11}P_1^c - b_{12}P_2^c - b_{13}P_3^c - b_{14}P_4^c - b_{15}P_5^c, \quad (31)$$

$$P_R(^{239}\mathrm{Pu(t)}) = P^c(^{239}\mathrm{Pu(t)}) - b_{21}P_1^c - b_{22}P_2^c - b_{23}P_3^c - b_{24}P_4^c - b_{25}P_5^c, \quad (32)$$

$$P_R(^{241}\mathrm{Pu(t)}) = P^c(^{241}\mathrm{Pu(t)}) - b_{31}P_1^c - b_{32}P_2^c - b_{33}P_3^c - b_{34}P_4^c - b_{35}P_5^c. \quad (33)$$

The superscript "c" denotes the summation calculation. The coefficients, b_{ij}, are calculated in the same way as Eq. (14). From their values in **Table 3**, we see that the decay heat powers of the thermal systems are nicely covered by those of the fast systems. It is remarkable that the decay heat of ^{239}Pu(t) is almost completely covered by that of the ^{239}Pu fast fission. It is also interesting that there is little cancellation due to negative coefficients in Eqs. (28) and (29), in which the linear combinations include the same fissiles with the different neutron energy.

Figure 3 shows the β and γ components of the decay heat powers obtained from Eqs. (28)–(30), together with the results of the ORNL measurements and summation calculations. Except for the γ decay heat of ^{235}U(t), there is reasonable agreement between the hybrid calculations and the ORNL measurements. A distinct deviation from the ORNL values is seen at about 2,000 s for the γ decay heat power of ^{235}U(t). It is reasonable to rule out the ORNL values at these cooling times because the summation calculation supports the hybrid calculation. Except for this case, the other ORNL values are reasonably consistent with the hybrid and summation calculations almost within the experimental uncertainties.

The residual terms, $P_R(^{235}\mathrm{U(t)})$ and $P_R(^{239}\mathrm{Pu(t)})$, reflect the neutron energy dependence of the decay heat power. In Fig. 3, the residual term is shown as the difference between the solid and dotted lines. Their values are 4–9% for ^{235}U(t) and 1–2% for ^{239}Pu(t). This implies that the neutron energy dependence is actually small but can be significant even for low energy neutron fissions.

From the analyses in the above two sections, we see the reasonable consistency among the decay heat measurements of the five fast (YAYOI) and three thermal (ORNL) fissioning systems between 20 and 4,000 s except for the γ decay heat of the ^{235}U thermal fission at about 2,000 s. In this way, the present method can be used to examine the consistency among the decay heat measurements even if the fissile nuclides and/or neutron energies are different.

V. Discussion

In the previous chapter, we see that the residual term P_R is relatively small, typically 10–20% when the fissile nuclides are different, and less than 10% when only the neutron energies are different. From this result, we may treat the residual power as a small correction term. However, this power is defined as a difference of the decay heat powers of different fissioning systems (Eqs. (23)–(27) and (31)–(33)), each of which is actually large. In the following, we examine the values of the residual term in more detail.

To begin with, we examine $\boldsymbol{y}_R$ values that are not used in the previous chapter. **Table 4** lists $|\boldsymbol{y}_R|/|\boldsymbol{y}|$ values of the five fast fissioning systems in the YAYOI measurement;

$$\boldsymbol{y}_R(^{232}\mathrm{Th}) = \boldsymbol{y}_1 - a_{12}\boldsymbol{y}_2 - a_{13}\boldsymbol{y}_3 - a_{14}\boldsymbol{y}_4 - a_{15}\boldsymbol{y}_5, \quad (34)$$

$$\boldsymbol{y}_R(^{233}\mathrm{U}) = \boldsymbol{y}_2 - a_{21}\boldsymbol{y}_1 - a_{23}\boldsymbol{y}_3 - a_{24}\boldsymbol{y}_4 - a_{25}\boldsymbol{y}_5, \quad (35)$$

$$\boldsymbol{y}_R(^{235}\mathrm{U}) = \boldsymbol{y}_3 - a_{31}\boldsymbol{y}_1 - a_{32}\boldsymbol{y}_2 - a_{34}\boldsymbol{y}_4 - a_{35}\boldsymbol{y}_5, \quad (36)$$

$$\boldsymbol{y}_R(^{238}\mathrm{U}) = \boldsymbol{y}_4 - a_{41}\boldsymbol{y}_1 - a_{42}\boldsymbol{y}_2 - a_{43}\boldsymbol{y}_3 - a_{45}\boldsymbol{y}_5, \quad (37)$$

$$\boldsymbol{y}_R(^{239}\mathrm{Pu}) = \boldsymbol{y}_5 - a_{51}\boldsymbol{y}_1 - a_{52}\boldsymbol{y}_2 - a_{53}\boldsymbol{y}_3 - a_{54}\boldsymbol{y}_4. \quad (38)$$

Table 3 Values of coefficients b_{ij} for three thermal fissioning systems of ORNL measurement in terms of five fast fissioning systems of YAYOI measurement

i (fissile (energy))	j (fissile)				
	1 (^{232}Th)	2 (^{233}U)	3 (^{235}U)	4 (^{238}U)	5 (^{239}Pu)
1 (^{235}U(t))	0.0620	0.2843	0.5963	0.1127	0.0224
2 (^{239}Pu(t))	−0.0222	0.0070	0.0194	0.0146	0.9754
3 (^{241}Pu(t))	−0.0522	−0.2618	0.1491	0.4221	0.7775

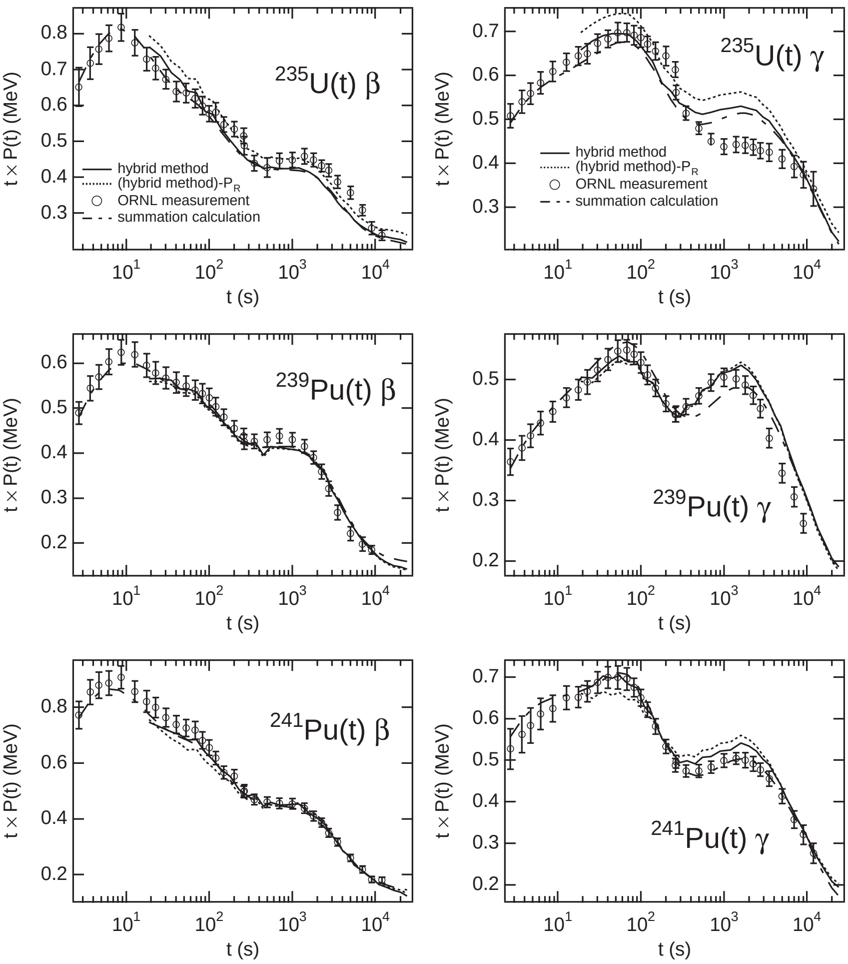

Fig. 3 Decay heat powers from pulse thermal fissions of ^{235}U, ^{239}Pu and ^{241}Pu

The solid lines show the $P(t)$ values obtained from Eqs. (28)–(30) while the dotted lines show the $P(t) - P_R(t)$ values. Also shown for comparison are the results of ORNL measurements (open circles with error bars) and summation calculations with ENDF/B-VI (dash-dotted lines).

From the $|\boldsymbol{y}_R|/|\boldsymbol{y}|$ value, we can estimate the portion of the fission yield which is not covered by the linear combination. It ranges from 0.27 to 0.54 and tends to be large for the heaviest and lightest fissiles. This suggests that the linear combination covers well the fission yields of the medium mass fissiles while the extrapolations to lighter or heavier fissiles become worse. Similarly, **Table 5** lists $|\boldsymbol{y}_R|/|\boldsymbol{y}|$ values of the three thermal fissioning systems in the ORNL measurements;

$$\boldsymbol{y}_R(^{235}\text{U(t)}) = \boldsymbol{y}(^{235}\text{U(t)}) - b_{11}\boldsymbol{y}_1 - b_{12}\boldsymbol{y}_2 - b_{13}\boldsymbol{y}_3 - b_{14}\boldsymbol{y}_4 - b_{15}\boldsymbol{y}_5, \quad (39)$$

$$\boldsymbol{y}_R(^{239}\text{Pu(t)}) = \boldsymbol{y}(^{239}\text{Pu(t)}) - b_{21}\boldsymbol{y}_1 - b_{22}\boldsymbol{y}_2 - b_{23}\boldsymbol{y}_3 - b_{24}\boldsymbol{y}_4 - b_{25}\boldsymbol{y}_5, \quad (40)$$

$$\boldsymbol{y}_R(^{241}\text{Pu(t)}) = \boldsymbol{y}(^{241}\text{Pu(t)}) - b_{31}\boldsymbol{y}_1 - b_{32}\boldsymbol{y}_2 - b_{33}\boldsymbol{y}_3 - b_{34}\boldsymbol{y}_4 - b_{35}\boldsymbol{y}_5. \quad (41)$$

The values for ^{235}U(t) and ^{239}Pu(t) reflect the difference from their fast systems. These values are much smaller, 0.17 for ^{235}U(t) and 0.12 for ^{239}Pu(t), than the others in Tables 4 and 5 that reflect the differences due to fissile nuclides. Nevertheless, one should be aware that the fission yield vector varies appreciably even with the neutron energy.

Table 4 The absolute values of residual yield vectors and their dominant components (FP's) among fast neutron fissions of YAYOI measurement

The FP's cover 50% of $|\boldsymbol{y}_R|$, and are listed in the descending order of importance. For those which are typed in bold, FP's in the same mass chains are listed in Table 6 for P_R.

Fissile	$\lvert\boldsymbol{y}_R\rvert/\lvert\boldsymbol{y}\rvert$	$\sum_k(\boldsymbol{y}_R)_k/2$	Dominant FP in $\boldsymbol{y}_R$
^{232}Th	0.5388	0.1379	^{137}Cs, **^{86}Se**, **^{87}Se**, **^{102}Zr**
^{233}U	0.3485	0.1246	^{88}Kr, ^{93}Sr, ^{89}Kr, ^{100}Zr, ^{92}Sr
^{235}U	0.2747	−0.1280	^{133}Te, ^{134}Te, ^{100}Zr, ^{132}Te, ^{96}Sr
^{238}U	0.4091	0.1581	^{103}Zr, ^{102}Zr, ^{100}Y, ^{135}Te, **^{136}Te**
^{239}Pu	0.4088	0.1230	^{104}Mo, ^{103}Mo

Table 5 The absolute values of residual yield vectors and their dominant components (FP's) for thermal fissions of ORNL measurements in terms of five fast fissions of YAYOI measurements

The FP's cover 50% of $|\boldsymbol{y}_R|$, and are listed in the descending order of importance. For those which are typed in bold, FP's in the same mass chains are listed in Tables 7–9 for P_R. The FP's with underscore are also listed in Tables 8 and 9.

Fissile (energy)	$\lvert\boldsymbol{y}_R\rvert/\lvert\boldsymbol{y}\rvert$	$\sum_k(\boldsymbol{y}_R)_k/2$	Dominant FP in $\boldsymbol{y}_R$
^{235}U(t)	0.1743	−0.0780	^{144}Ba, ^{133}Te, ^{92}Sr, ^{140}Cs, **^{134}Te**, ^{100}Zr
^{239}Pu(t)	0.1239	0.0059	**^{132}Sb**, **^{132}Te**, **^{140}Ba**, **^{136}Xe**
^{241}Pu(t)	0.2513	−0.0350	**^{108}Tc**, **^{106}Mo**, **^{107}Mo**, **^{138}Xe**

Then, why is P_R/P much smaller than $|\boldsymbol{y}_R|/|\boldsymbol{y}|$? This stems from cancellations among FP contributions to P_R because the components of $\boldsymbol{y}_R$ can be negative. Actually, the sum of all components of $\boldsymbol{y}_R$ is sufficiently small. This value can be calculated from the coefficients of the linear combination. For ^{232}Th, it is given by

$$\sum_k(\boldsymbol{y}_R)_k/2 = 1 - a_{12} - a_{13} - a_{14} - a_{15}. \tag{42}$$

Here, we consider half of the sum for convenience because the sum of the independent fission yields is normalized to be two in ENDF/B-VI. As listed in Tables 4 and 5, the values of $\sum_k(\boldsymbol{y}_R)_k/2$ are much smaller than the $|\boldsymbol{y}_R|/|\boldsymbol{y}|$ values. This implies the smallness of P_R/P values, that are differences between the solid and dotted lines in Figs. 2 and 3.

In fact, there are substantial cancellations among FP contributions in P_R. Dominant FP's in P_R at typical cooling times are listed in **Tables 6–9** while those in $\boldsymbol{y}_R$ are listed in Tables 4 and 5, too. The FP's which appear in both the P_R and $\boldsymbol{y}_R$ tables are typed in bold with underscore. Except for ^{235}U and ^{238}U in Table 6 and ^{235}U(t) in Table 7, we see from these tables that the smallness of $|P_R|$ is a consequence of cancellations among individual contributions with different signs. It is also noteworthy that the dominant FP's in $\boldsymbol{y}_R$ do not always give dominant contributions to P_R due to the smallness of their decay data. In fact, most of the FP's and their mass chains in the $\boldsymbol{y}_R$ tables do not appear in the P_R tables.

It is more difficult to evaluate uncertainties in hybrid calculations than in usual summation calculations because the key formula (15) has complicated dependence on fission yield vectors of five fissioning systems. Here, we give a brief qualitative discussion of the uncertainties in hybrid calculations. The uncertainties stem from three kinds of input data; measured decay heat powers, FP fission yields and FP decay data. We believe that the uncertainties primarily stem from measured decay heat powers and secondly from fission yields because of the following two observations.

(i) The uncertainties from the FP decay data are expected to be much smaller than those in usual summation calculations. A rough estimate of this ratio is $|\boldsymbol{y}_R|/|\boldsymbol{y}|$, which is usually less than 1/2 as shown in Tables 4 and 5. In addition, the strong cancellations among the FP decay heat contributions work to reduce the uncertainties in hybrid calculations. If we notice that the uncertainty from the decay data is only 1–2% in usual summation calculations even for minor actinides,[10–12] the one in hybrid calculations would be negligibly small.

(ii) Uncertainties in the FP fission yield data can affect hybrid calculations through P_R and the coefficients of the linear combination. From the smallness of the P_R values, we may also neglect the uncertainties in P_R. Then, the primary uncertainties from the fission yields are brought about through the coefficients of the linear combination. However, detailed behaviors of the fission yields are not very important because these coefficients reflect gross properties of the fission yields. Therefore, for the fissioning systems in the previous chapter whose fission yields are relatively well known, the uncertainties due to the fission yields are expected to be small.

We can also calculate the decay heat of minor actinides in the hybrid method. Because their fission yields are not precisely known, the prediction power will not be so good as the one for the cases in the previous chapter. However, the present method still has an advantage over the usual summation calculation because we can utilize the true decay data values implicitly contained in the measured decay heat values. Therefore, with the present method, we could improve the decay heat evaluation of any fissioning systems even if no measurement has been performed so far for the systems.

VI. Conclusion

We propose a hybrid method for calculating the aggregate decay heat power making full use of its macroscopic-measurement data. The term hybrid means that this method bridges the macroscopic measurements and summation calculations. The method is derived from the linearity of the decay heat power to the fission yields and based on an exact mathematical relation among decay heat powers of different fissioning systems.

The hybrid method enables us to calculate an aggregate decay heat power of a fissioning system from measured decay heat powers of other fissioning systems. The method is first applied to analyze the consistency among the decay heat measurements at YAYOI of the fast neutron induced fissions of ^{232}Th, ^{233}U, ^{235}U, ^{238}U and ^{239}Pu. Specifically, the de-

Table 6 The dominant FP's in P_R in Eqs. (23)–(27)

The absolute values of their individual contributions to P_R from the listed FP's are larger than 20% of P_R. The FP's are listed in the descending order of importance. For those which are typed in bold, FP's in the same mass chains are listed in Table 4 for $\boldsymbol{y}_R$.

	t (s)	β component		γ component	
		P_R/P (%)	Dominant FP in P_R	P_R/P (%)	Dominant FP in P_R
Eq. (23) (^{232}Th)	19	15.39	^{88}Br, ^{100}Nb	26.17	
	110	22.57	**^{86}Br**	25.90	**^{86}Br**, **^{87}Br**
	1,200	10.25	**^{102}Tc**, ^{89}Rb, ^{84}Br, ^{139}Cs	7.795	^{89}Rb, ^{101}Mo, ^{84}Br, **^{102}Tc**
	11,000	26.84	^{88}Rb, **^{87}Kr**	13.47	^{88}Kr, ^{142}La, ^{134}I, **^{87}Kr**
Eq. (24) (^{233}U)	19	2.633	^{135}Te, ^{88}Br, ^{137}I, **^{100}Nb**, ^{104}Nb, ^{96}Y, ^{136}Te, ^{91}Rb, ^{134m}Sb, ^{96m}Y	4.996	^{96m}Y, ^{88}Br, ^{104}Nb, ^{136}Te, ^{91}Rb
	110	7.781	^{91}Rb	9.323	^{91}Rb
	1,200	10.83	^{94}Y	13.34	^{89}Rb
	11,000	17.02	^{88}Rb	17.12	
Eq. (25) (^{235}U)	19	−7.749		−12.57	
	110	−12.75		−12.61	
	1,200	−10.33		−10.68	^{104}Tc
	11,000	−11.57		−9.053	
Eq. (26) (^{238}U)	19	15.74		11.55	
	110	12.43		10.82	^{86}Br, **^{136}I**, ^{87}Br
	1,200	15.77		18.16	
	11,000	6.183	^{88}Rb, ^{87}Kr, ^{134}I	14.31	^{134}I
Eq. (27) (^{239}Pu)	19	8.745	^{106}Tc	15.20	^{106}Tc
	110	15.92		13.33	^{106}Tc
	1,200	11.49	**^{104}Tc**, ^{105}Tc	8.365	**^{104}Tc**, ^{89}Rb
	11,000	−2.123	^{88}Rb, ^{92}Y, ^{105}Ru, ^{93}Y, ^{139}Ba, ^{91}Sr, ^{141}La, ^{88}Kr, ^{92}Sr, ^{145}Pr, ^{142}La	0.3091	^{92}Sr, ^{105}Ru, ^{88}Kr, ^{135}I, ^{91}Sr, ^{142}La, ^{134}I, ^{88}Rb, ^{133m}Te, ^{138}Cs, ^{91m}Y, ^{92}Y, **^{104}Tc**, ^{84}Br, ^{135}Xe, ^{149}Nd, ^{87}Kr, ^{146}Pr, ^{133}I, ^{133}Te, ^{134}Te, ^{128m}Sb, ^{93}Y

Table 7 The dominant FP's in P_R for ^{235}U(t) in Eq. (31)

The absolute values of their individual contributions to P_R from the listed FP's are larger than 20% of P_R. The FP's are listed in the descending order of importance. For those which are typed in bold, FP's in the same mass chains are also listed in Tables 5 for $\boldsymbol{y}_R$.

^{235}U(t) t (s)	β component		γ component	
	P_R/P (%)	Dominant FP in P_R	P_R/P (%)	Dominant FP in P_R
19	−4.009		−5.982	
110	−6.667		−6.963	
1,200	−6.299		−6.318	
11,000	−8.609	^{88}Rb	−4.668	**^{134}I**, ^{88}Kr

cay heat power of one of these fissioning systems is calculated from the rest in the hybrid method. The method works well in these calculations. Typically, 80–90% of the decay heat power is covered by the measured values. The results obtained in the hybrid method agree well with the measured powers before 4,000 s. Furthermore, the present results are also consistent with the summation calculations. Therefore, we conclude that the decay heat evaluations of the five systems in the three methods are reliable before 4,000 s while there is inconsistency after 4,000 s due to an unknown reason.

The present method also works well for systems with different neutron energies. We examine the decay heat powers of the thermal neutron fissions of ^{235}U, ^{239}Pu and ^{241}Pu measured at ORNL, using the YAYOI values of the five fast fissioning systems. The decay heat powers of these thermal fissioning systems are covered well by the fast fissioning systems. The hybrid as well as summation calculations agree well with the ORNL values, except that the ORNL values distinctively differ from these two calculations for the γ decay heat of the ^{235}U thermal fission at about 2,000 s. Therefore, we can conclude reliability of the decay heat evaluations for the five fast and three thermal systems before 4,000 s, except for the γ decay heat measurement of the ^{235}U thermal fission at about 2,000 s.

We can improve the decay heat evaluations by accumulating consistent decay heat measurements using the hybrid method. This method works best when fissioning systems have similar fission yields. Therefore, from the decay heat measurements for minor actinide fissiles being performed by Japan Nuclear Cycle Development Institute,[13] we will be able to improve the decay heat evaluation in the wide range of the fissile.

The effectiveness of the hybrid method essentially depends on the smallness of the residual term P_R due to strong can-

Table 8 The dominant FP's in P_R for ^{239}Pu(t) in Eq. (32)

The absolute values of their individual contributions to P_R from the listed FP's are larger than 20% of P_R. The FP's are listed in the descending order of importance. For those which are typed in bold, FP's in the same mass chains are listed in Tables 5 for $\boldsymbol{y}_R$. The FP's in bold with underscore are also listed in Table 5 for $\boldsymbol{y}_R$.

^{239}Pu(t)	β component		γ component	
t (s)	P_R/P (%)	Dominant FP in P_R	P_R/P (%)	Dominant FP in P_R
19	0.9400	^{137}I, ^{101}Nb, ^{100}Nb, ^{96}Y, ^{138}I, ^{141}Cs, ^{91}Kr, ^{140}Cs, **^{136}I**, ^{105}Mo, ^{109}Ru	1.314	^{96m}Y, ^{138}I, **^{140}Cs**, ^{137}I, **^{136}I**
110	0.8260	**^{136}I**, **^{140}Cs**, **^{132}Sb**, ^{109}Rh, ^{137}Xe, ^{105}Mo, ^{144}La, ^{137}I, ^{109}Ru, ^{91}Rb, ^{102}Tc	0.4800	**^{132}Sb**, **^{136}I**, **^{140}Cs**, ^{144}La, ^{91}Rb, ^{106}Tc, ^{86}Br, ^{99m}Nb, ^{148m}Pr, ^{132}Sn, ^{105}Mo, ^{132m}Sb, ^{101}Mo, ^{102}Tc, ^{109}Ru, ^{104}Mo, ^{90m}Rb, ^{137}I, ^{94}Sr, ^{108}Rh, ^{109}Rh, ^{148}Pr, ^{133}Te, ^{129}Sn, ^{104}Tc, ^{130m}Sb, ^{141}Cs, ^{98}Nb
1,200	−0.4016	^{102}Tc, ^{104}Tc, ^{105}Tc, ^{101}Mo, ^{132}Sb, ^{101}Tc, ^{133}Te, ^{138}Xe, ^{102}Mo, ^{131}Sb, ^{137}Xe, ^{130m}Sb, ^{143}La, ^{141}Ba, ^{95}Y, ^{139}Cs, ^{108}Rh, ^{94}Y, ^{134}I	−0.7958	^{101}Mo, ^{104}Tc, ^{102}Tc, **^{132}Sb**, ^{131}Sb, ^{133}Te, ^{138}Xe, ^{130m}Sb, ^{134}I, ^{105}Tc, ^{133m}Te, ^{101}Tc
11,000	1.364	^{142}La, ^{128m}Sb, ^{141}La	1.781	^{142}La, ^{135}I, ^{134}I

Table 9 The dominant FP's in P_R for ^{241}Pu(t) in Eq. (33)

The absolute values of their individual contributions to P_R from the listed FP's are larger than 20% of P_R. The FP's are listed in the descending order of importance. For those which are typed in bold, FP's in the same mass chains are listed in Tables 5 for $\boldsymbol{y}_R$. The FP's in bold with underscore are also listed in Table 5 for $\boldsymbol{y}_R$.

^{241}Pu(t)	β component		γ component	
t (s)	P_R/P (%)	Dominant FP in P_R	P_R/P (%)	Dominant FP in P_R
19	−1.419	^{100}Nb, ^{96}Y, **^{108}Tc**, **^{107}Tc**, **^{106}Tc**, **^{106}Mo**, ^{92}Rb, ^{93}Rb, ^{101}Nb, ^{91}Kr, ^{98}Nb, ^{109}Ru, ^{95}Sr, ^{105}Mo, ^{100}Zr, ^{139}Xe, ^{98}Zr, ^{99}Nb, ^{143}Ba, ^{137}I, **^{107}Mo**	2.827	**^{108}Tc**, **^{106}Tc**, **^{107}Tc**, ^{95}Sr, ^{91}Kr, ^{98}Nb, **^{106}Mo**, ^{110m}Rh, ^{96}Y
110	4.620	**^{106}Tc**, ^{109}Rh, **^{108}Rh**	3.151	**^{106}Tc**, ^{110m}Rh, ^{91}Rb, ^{108}Rh, ^{132m}Sb, ^{103}Mo, ^{140}Cs
1,200	−1.172	^{102}Tc, ^{94}Y, ^{95}Y, **^{108}Rh**, **^{107}Rh**, ^{139}Cs, ^{101}Mo, **^{138}Xe**, ^{101}Tc, **^{107}Ru**, ^{93}Sr, **^{108}Ru**, ^{102}Mo, ^{105}Tc	−3.804	^{101}Mo, ^{93}Sr, ^{102}Tc, ^{95}Y, **^{138}Xe**
11,000	−0.7296	^{92}Y, ^{88}Rb, ^{93}Y, ^{139}Ba, ^{138}Cs, ^{87}Kr, ^{142}La, ^{109}Pd, ^{97}Zr, ^{91}Sr, ^{141}La, ^{129}Te, ^{131}Te, ^{128m}Sb, ^{129}Sb, ^{92}Sr, ^{97}Nb, ^{135}Xe, ^{88}Kr, ^{105}Ru, ^{134}I, ^{133}I	−0.4813	^{92}Sr, ^{138}Cs, ^{142}La, ^{88}Kr, ^{129}Sb, ^{134}I, ^{128m}Sb, ^{91}Sr,^{97m}Nb, ^{133m}Te, ^{130}Sb, ^{88}Rb, ^{87}Kr, ^{97}Nb, ^{105}Ru, ^{133}I, ^{92}Y, ^{134}Te, ^{91m}Y,^{135}Xe, ^{131}Te, ^{127}Sn

cellations among individual FP contributions. Consequently, the decay data is minimally used in this method. Actually, we see significant cancellations among components of the residual yield vector $\boldsymbol{y}_R$ although the absolute value of the residual vector is relatively large.

The present method is derived from a mathematical relation among the fission yields of different fissioning systems. The fission yield is the only quantity that characterizes a fissioning system. Therefore, it is highly desirable to perform precise determination of the fission yields systematically in the wide range of the fissile. Unfortunately, the present evaluations are largely dependent on systematics.

Uncertainties in hybrid calculations are more complicated than those for usual summation calculations. In this paper, we demonstrate only a qualitative argument. However, further study is necessary to use this method in more precise quantitative analyses.

Lastly, we note that the hybrid method is also applicable to other aggregate FP properties, such as the average delayed neutron yield ($\bar{\nu}_d$), and its time dependence. So far we have applied the hybrid method to them and obtained a preliminary result. The detailed analyses for the delayed neutron properties will be reported in a separate paper.

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Appendix

Derivation of Eq. (14)

The coefficient values of the linear combination in Eq. (13) is chosen to minimize

$$|\boldsymbol{y}_R|^2 = |\boldsymbol{y} - a_1\boldsymbol{y}_1 - a_2\boldsymbol{y}_2 - a_3\boldsymbol{y}_3 - a_4\boldsymbol{y}_4|^2. \tag{A1}$$

From the condition that $|\boldsymbol{y}_R|^2$ is minimum against the variation of a_i, we have

$$\frac{\partial|\boldsymbol{y}_R|^2}{\partial a_i} = 2\left(\sum_{j=1}^{4} a_j\boldsymbol{y}_i\cdot\boldsymbol{y}_j - \boldsymbol{y}_i\cdot\boldsymbol{y}\right) = 0. \tag{A2}$$

This condition can be written in a compact form in the matrix representation;

$$\begin{pmatrix} \boldsymbol{y}_1\cdot\boldsymbol{y}_1 & \boldsymbol{y}_1\cdot\boldsymbol{y}_2 & \boldsymbol{y}_1\cdot\boldsymbol{y}_3 & \boldsymbol{y}_1\cdot\boldsymbol{y}_4 \\ \boldsymbol{y}_2\cdot\boldsymbol{y}_1 & \boldsymbol{y}_2\cdot\boldsymbol{y}_2 & \boldsymbol{y}_2\cdot\boldsymbol{y}_3 & \boldsymbol{y}_2\cdot\boldsymbol{y}_4 \\ \boldsymbol{y}_3\cdot\boldsymbol{y}_1 & \boldsymbol{y}_3\cdot\boldsymbol{y}_2 & \boldsymbol{y}_3\cdot\boldsymbol{y}_3 & \boldsymbol{y}_3\cdot\boldsymbol{y}_4 \\ \boldsymbol{y}_4\cdot\boldsymbol{y}_1 & \boldsymbol{y}_4\cdot\boldsymbol{y}_2 & \boldsymbol{y}_4\cdot\boldsymbol{y}_3 & \boldsymbol{y}_4\cdot\boldsymbol{y}_4 \end{pmatrix} \begin{pmatrix} a_1 \\ a_2 \\ a_3 \\ a_4 \end{pmatrix} = \begin{pmatrix} \boldsymbol{y}_1\cdot\boldsymbol{y} \\ \boldsymbol{y}_2\cdot\boldsymbol{y} \\ \boldsymbol{y}_3\cdot\boldsymbol{y} \\ \boldsymbol{y}_4\cdot\boldsymbol{y} \end{pmatrix}. \tag{A3}$$

Then, by multiplying the inverse of the matrix from the left side, we finally obtain Eq. (14).