Decay Heat Calculation for Minor Actinides in the Hybrid Method

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For evaluation of decay heat from the fission of minor actinides (MAs) we are currently forced to rely only on summation calculations since there has been no direct measurement for these nuclides. In this paper, we use an alternative approach (named “hybrid” method) to evaluate the MA decay heat power using the measured decay heat powers for major actinides at YAYOI. Results for the fast fissions of ^{237}Np, ^{241}Am, ^{243}Am, ^{242}Cm and ^{244}Cm are in support of the summation calculations for β decay heat, but suggest notable uncertainties in γ decay-heat summation calculations.

1. Introduction

There is a growing need for accurating evaluation of decay heat from the fission of minor actinides (MA) since increasing efforts are being made for realization of MA burner. However, at present only summation calculations are available for this purpose because no direct measurement has been performed for these nuclides. In this paper, decay heat powers from the fast fissions of ^{237}Np, ^{241}Am, ^{243}Am, ^{242}Cm and ^{244}Cm are calculated in an alternative approach, called hybrid method[1, 2], in which we utilize measured decay heat powers of Th, U and Pu isotopes, to examine the reliability of the summation calculations.

2. Hybrid method

The method used in this paper is based on the fact that the decay heat power is a linear function of the independent fission yield. In the matrix representation of the summation method, we can express the decay heat power , P, with a vector of independent fission yield, $\boldsymbol{y}$, as

$$P=\boldsymbol{E}\boldsymbol{\Lambda}_0\exp(\Lambda t)\boldsymbol{y}, \quad (1)$$

$$\exp(\Lambda t) = I + \Lambda t + \frac{1}{2!}(\Lambda t)^2 + \cdots, \quad (I: \ \text{unit matrix}) \quad (2)$$

where $\boldsymbol{\Lambda}$ and $\boldsymbol{\Lambda_0}$ represent matrices with decay constants,

$$\Lambda=\begin{pmatrix} -\lambda^1 & b^{2\to1}\lambda^2 & \cdots & b^{N\to1}\lambda^N \\ b^{1\to2}\lambda^1 & -\lambda^2 & \cdots & b^{N\to2}\lambda^N \\ \vdots & \vdots & \ddots & \vdots \end{pmatrix} \quad \text{and} \quad \Lambda^0=\begin{pmatrix} -\lambda^1 & 0 & \cdots & 0 \\ 0 & -\lambda^2 & \cdots & 0 \\ \vdots & \vdots & \ddots & \vdots \end{pmatrix}, \quad (3)$$

respectively. In these matrices, λ_k and $b_{j\to k}$ are the decay constant of nuclei k, and branching ratio from nuclei j to nuclei k, respectively. The vector $\boldsymbol{E}$ is the average decay energy vector defined as

$$\boldsymbol{E} \quad E_1 \quad E_2 \quad E_N \quad (4)$$

In the hybrid method, we start with writing $\boldsymbol{y}$ for a given fissioning system as a linear combination of yield vectors of other N fissioning systems, $\boldsymbol{y}_1\cdots\boldsymbol{y}_N$,

$$\boldsymbol{y}=a_1\boldsymbol{y}_1+a_2\boldsymbol{y}_2+\cdots+a_N\boldsymbol{y}_N+\boldsymbol{y}_R, \quad (5)$$

where $\boldsymbol{y}_R$ is the residual vector. The values of the coefficients, a_i, are chosen to minimize $|\boldsymbol{y}_R|$,

$$
|\boldsymbol{y}_R| = \min \quad \Leftrightarrow \quad \begin{cases} \boldsymbol{y}\cdot\boldsymbol{y}_1 = a_1|\boldsymbol{y}_1|^2 + a_2\boldsymbol{y}_2\cdot\boldsymbol{y}_1 + \cdots + a_N\boldsymbol{y}_N\cdot\boldsymbol{y}_1 \\ \boldsymbol{y}\cdot\boldsymbol{y}_2 = a_1\boldsymbol{y}_1\cdot\boldsymbol{y}_2 + a_2|\boldsymbol{y}_2|^2 + \cdots + a_N\boldsymbol{y}_N\cdot\boldsymbol{y}_2 \\ \vdots \\ \boldsymbol{y}\cdot\boldsymbol{y}_N = a_1\boldsymbol{y}_1\cdot\boldsymbol{y}_N + a_2\boldsymbol{y}_2\cdot\boldsymbol{y}_N + \cdots + a_N|\boldsymbol{y}_N|^2 . \end{cases} \tag{6}
$$

so that,

$$
\begin{pmatrix} a_1 \\ a_2 \\ \vdots \\ a_N \end{pmatrix} = \begin{pmatrix} |\boldsymbol{y}_1|^2 & \boldsymbol{y}_2\cdot\boldsymbol{y}_1 & \cdots & \boldsymbol{y}_N\cdot\boldsymbol{y}_1 \\ \boldsymbol{y}_1\cdot\boldsymbol{y}_2 & |\boldsymbol{y}_2|^2 & \cdots & \boldsymbol{y}_N\cdot\boldsymbol{y}_2 \\ \vdots & \vdots & \ddots & \vdots \\ \boldsymbol{y}_1\cdot\boldsymbol{y}_N & \boldsymbol{y}_2\cdot\boldsymbol{y}_N & \cdots & |\boldsymbol{y}_N|^2 \end{pmatrix}^{-1} \begin{pmatrix} \boldsymbol{y}\cdot\boldsymbol{y}_1 \\ \boldsymbol{y}\cdot\boldsymbol{y}_2 \\ \vdots \\ \boldsymbol{y}\cdot\boldsymbol{y}_N \end{pmatrix}. \tag{7}
$$

The above-mentioned linearity leads to the following expression for the decay heat power, P, with the same coefficients, a_i, as obtained from (7),

$$
P = a_1P_1 + a_2P_2 + \cdots + a_NP_N + P_R. \tag{8}
$$

Here, P_i denotes the decay heat power of fissioning system i. The residual term P_R can be calculated in the summation method using the yield vector $\boldsymbol{y}_R$. Note that P_R is expected to be small because $|\boldsymbol{y}_R|$ has been minimized. We thus can evaluate P for such nuclides as MAs, for which no reliable measurement data exist, by using integral measurement data P_i for other fissioning systems.

We summarize the nuclear properties required for decay power evaluation with the hybrid method, the summation calculation and the integral measurement in Table 1. The accuracy of the summation calculation fully depends on the accuracy of the decay data whilst the hybrid method uses decay data only in the calculation of the small residual term P_R.

3. Results

The decay heat powers of ^{237}Np(F), ^{241}Am(F), ^{243}Am(F), ^{242}Cm(F) and ^{244}Cm(F) have been calculated in the hybrid method using the decay heat powers of ^{232}Th(F), ^{233}U(F), ^{235}U(F), ^{238}U(F), ^{239}Pu(F) measured at YAYOI. Here, the symbol "F" stands for the fast fission. We get the following five equations from Eq. (8),

$$
P(^{237}\mathrm{Np}) = a_1P(^{232}\mathrm{Th}) + a_2P(^{233}\mathrm{U}) + a_3P(^{235}\mathrm{U}) + a_4P(^{238}\mathrm{U}) + a_5P(^{239}\mathrm{Pu}) + P_R(^{237}\mathrm{Np}), \tag{9}
$$

$$
P(^{241}\mathrm{Am}) = b_1P(^{232}\mathrm{Th}) + b_2P(^{233}\mathrm{U}) + b_3P(^{235}\mathrm{U}) + b_4P(^{238}\mathrm{U}) + b_5P(^{239}\mathrm{Pu}) + P_R(^{241}\mathrm{Am}), \tag{10}
$$

$$
P(^{243}\mathrm{Am}) = c_1P(^{232}\mathrm{Th}) + c_2P(^{233}\mathrm{U}) + c_3P(^{235}\mathrm{U}) + c_4P(^{238}\mathrm{U}) + c_5P(^{239}\mathrm{Pu}) + P_R(^{243}\mathrm{Am}), \tag{11}
$$

$$
P(^{242}\mathrm{Cm}) = d_1P(^{232}\mathrm{Th}) + d_2P(^{233}\mathrm{U}) + d_3P(^{235}\mathrm{U}) + d_4P(^{238}\mathrm{U}) + d_5P(^{239}\mathrm{Pu}) + P_R(^{242}\mathrm{Cm}), \tag{12}
$$

$$
P(^{244}\mathrm{Cm}) = e_1P(^{232}\mathrm{Th}) + e_2P(^{233}\mathrm{U}) + e_3P(^{235}\mathrm{U}) + e_4P(^{238}\mathrm{U}) + e_5P(^{239}\mathrm{Pu}) + P_R(^{244}\mathrm{Cm}). \tag{13}
$$

The fission yields and decay data of ENDF/B-VI have been used to calculate the values of coefficients a_i, b_i, c_i, d_i and e_i from Eq. (7) and then to obtain $\boldsymbol{y}_R$ and P_R. As shown in Table 2, the coefficients are all positive for ^{237}Np(F), but take also negative values for ^{241}Am(F), ^{243}Am(F), ^{242}Cm(F) and ^{244}Cm(F). We see from Table 2 that the offsets due to the negative terms are significant.

The decay heat power from ^{237}Np(F), ^{241}Am(F), ^{243}Am(F), ^{242}Cm(F) and ^{244}Cm(F) are shown in Figs. 1 through 5. The linear combination of measurement data, in the right hand side of Eqs. (9) through (13), amounts to about 90% of the total estimated decay power for each fissioning system as listed in Tables 3 through 7. Therefore the present results could be taken as almost independent of summation calculations. The present results are compared with the summation calculations in Fig. 6. For β decay heat, the hybrid and summation calculations agree reasonably within the uncertainties in summation calculations (3~5 % [4]). However, for γ decay heat the hybrid calculations differ appreciably from the summation calculations beyond their uncertainties (4~8 % [4]).

4. Conclusion

The decay heat powers from the fast fissions of ^{237}Np, ^{241}Am, ^{243}Am, ^{242}Cm and ^{244}Cm have been estimated from the measured decay heat powers from the fast fissions of Th, U and Pu isotopes. The fact that the decay heat power is a linear function of the independent fission yield was used to estimate the minor actinides decay heat powers, for which there exist no data, from data for other fissioning systems. The results have been used to examine the reliability of summation calculations for these minor actinide fissioning systems. Although the comparisons are favorable for the β decay heat summation calculations, they suggest that further confirmation should be done for the γ decay heat summation calculations.

References

[1] M. Akiyama et al., J. Atom. Enter. Soc. of Japan, 24, 709(1982)
[2] M. Akiyama et al., J. Atom. Enter. Soc. of Japan, 24, 803(1982)
[3] M. Akiyama, Proc. Int. Conf. on Nucl. Data for Sci. and Tech., Antwerp, 1982, p.273.
[4] H. Ohta et al., Proc. 1995 Symposium on Nuclear Data, JAERI-Conf 96-008, p.p.290-295.

Table 1. Nuclear properties required for calculation (●:required, △:partly required, ×:not required).

Nuclear properties	Summation method	New method	Integral measurement
Independent fission yields	●	●	×
Decay constants	●	△	×
Branching ratios	●	△	×
Average decay energies	●	△	×

Table 2. Values of coefficients a_i, b_i, c_i, d_i and e_i.

Coefficients	1	2	3	4	5
a_i	0.005	0.069	0.274	0.081	0.557
b_i	0.042	0.040	-0.180	-0.128	1.122
c_i	-0.002	-0.129	-0.006	0.061	0.995
d_i	0.107	0.314	-0.648	-0.162	1.140
e_i	0.079	-0.085	-0.170	-0.152	1.190

Table 3. Values of $[a_1P(^{232}\mathrm{Th})+a_2P(^{233}\mathrm{U})+a_3P(^{235}\mathrm{U})+a_4P(^{238}\mathrm{U})+a_5P(^{239}\mathrm{Pu})]/P(^{237}\mathrm{Np})$.

t(s)	β decay heat	γ decay heat
90	97.6 %	97.1 %
900	99.2 %	97.8 %
9000	98.6 %	99.4 %

Table 4. Values of $[b_1P(^{232}\mathrm{Th})+b_2P(^{233}\mathrm{U})+b_3P(^{235}\mathrm{U})+b_4P(^{238}\mathrm{U})+b_5P(^{239}\mathrm{Pu})]/P(^{241}\mathrm{Am})$.

t(s)	β decay heat	γ decay heat
90	89.9 %	89.9 %
900	89.4 %	93.3 %
9000	88.1 %	90.8 %

Table 5. Values of [$c_1P(^{232}\mathrm{Th})+c_2P(^{233}\mathrm{U})+c_3P(^{235}\mathrm{U})+c_4P(^{238}\mathrm{U})+c_5P(^{239}\mathrm{Pu})$]/ $P(^{243}\mathrm{Am})$.

t(s)	β decay heat	γ decay heat
90	86.4 %	86.8 %
900	91.0 %	98.0 %
9000	93.9 %	99.9 %

Table 6. Values of [$d_1P(^{232}\mathrm{Th})+d_2P(^{233}\mathrm{U})+d_3P(^{235}\mathrm{U})+d_4P(^{238}\mathrm{U})+d_5P(^{239}\mathrm{Pu})$]/ $P(^{242}\mathrm{Cm})$.

t(s)	β decay heat	γ decay heat
90	91.5 %	92.1 %
900	82.6 %	88.5 %
9000	87.0 %	92.9 %

Table 7. Values of [$e_1P(^{232}\mathrm{Th})+e_2P(^{233}\mathrm{U})+e_3P(^{235}\mathrm{U})+e_4P(^{238}\mathrm{U})+e_5P(^{239}\mathrm{Pu})$]/ $P(^{244}\mathrm{Cm})$.

t(s)	β decay heat	γ decay heat
90	84.0 %	86.1 %
900	84.4 %	93.8 %
9000	89.1 %	104.7 %

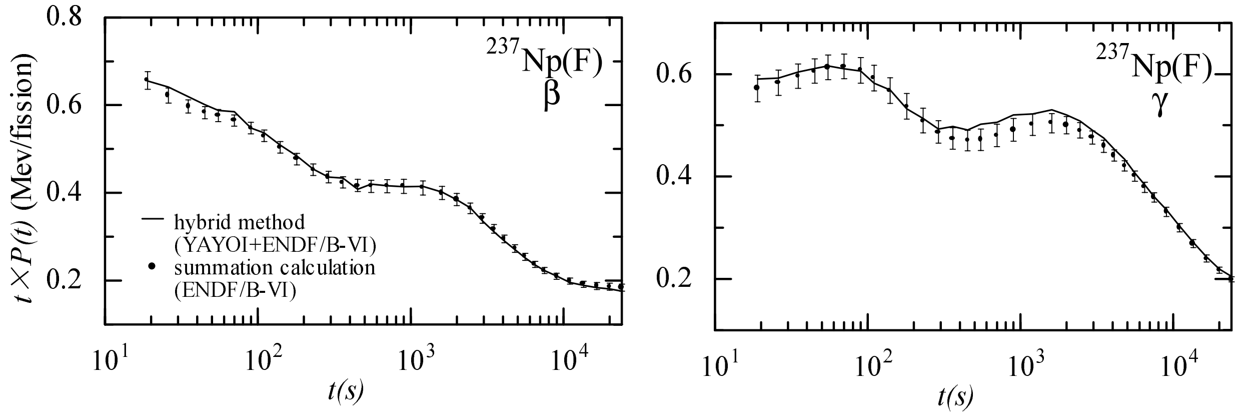

Fig. 1. The β and γ decay heat powers of ^{237}Np in the hybrid method (solid line) and in the summation method (filled circles with error bars).

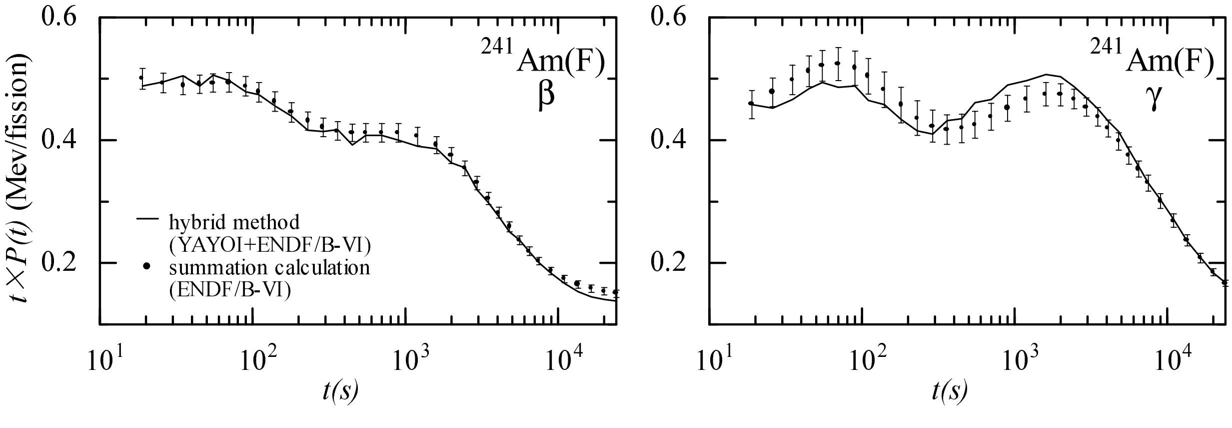

Fig. 2. The β and γ decay heat powers of ^{241}Am in the hybrid method (solid line) and in the summation method (filled circles with error bars).

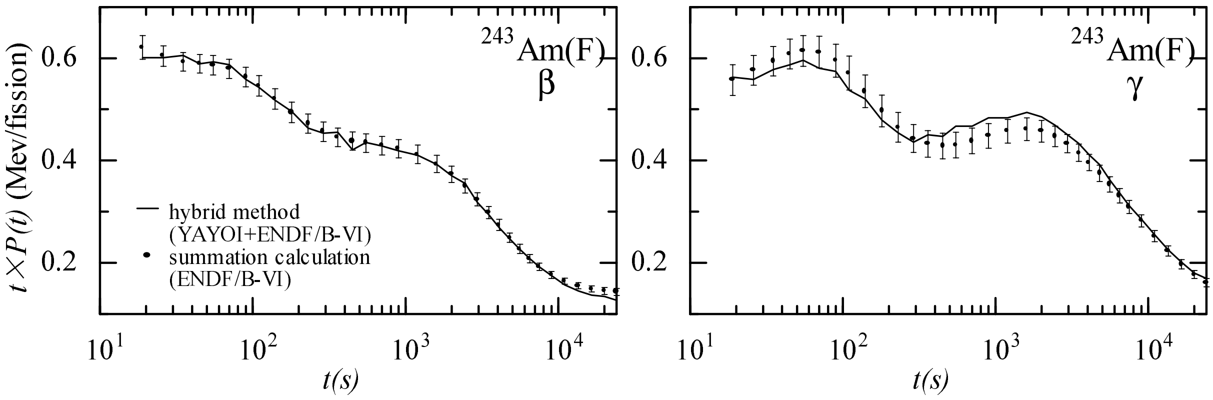

Fig. 3. The β and γ decay heat powers of ^{243}Am in the hybrid method (solid line) and in the summation method (filled circles with error bars).

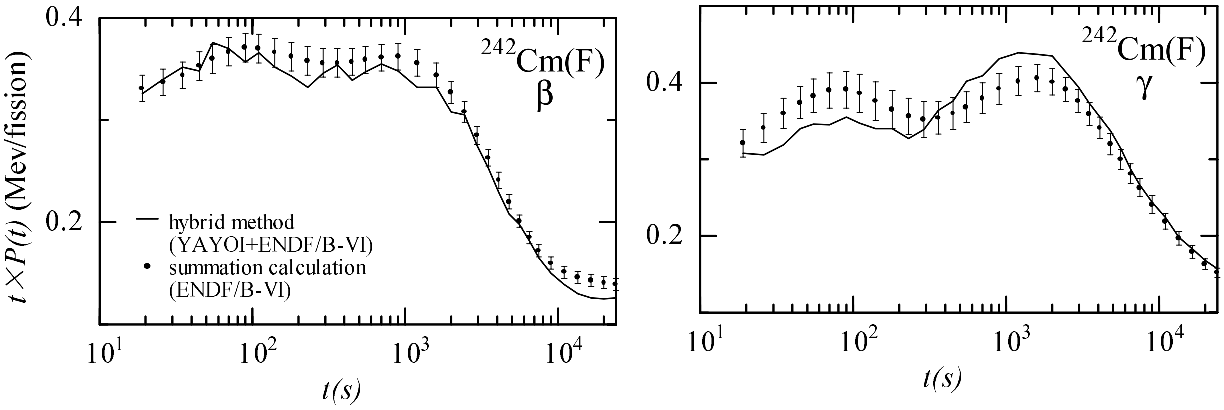

Fig. 4. The β and γ decay heat powers of ^{242}Cm in the hybrid method (solid line) and in the summation method (filled circles with error bars).

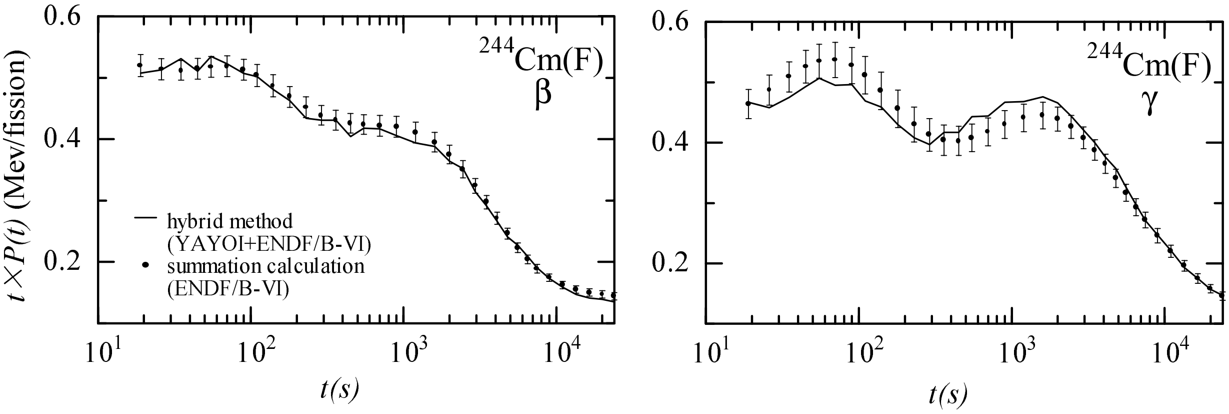

Fig. 5. The β and γ decay heat powers of ^{244}Cm in the hybrid method (solid line) and in the summation method (filled circles with error bars).

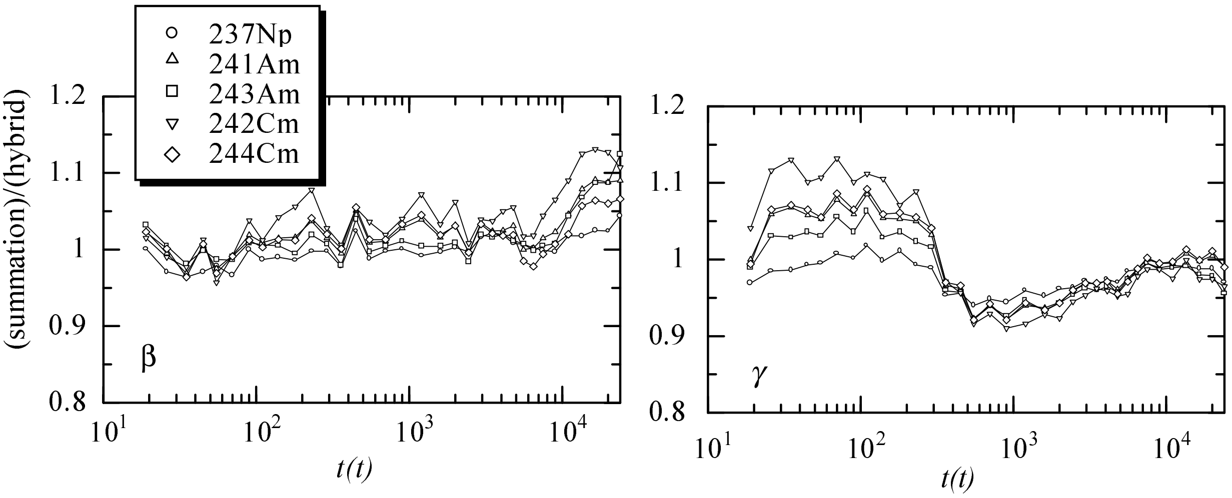

Fig. 6. Comparison between the summation and hybrid calculations for ^{237}Np, ^{241}Am, ^{243}Am, ^{242}Cm and ^{244}Cm.