Simplified Sensitivity Method for Uncertainty Evaluation in Decay-Heat Calculations due to Experimental Errors of Fission Yields

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A simplified method to evaluate uncertainty of calculated decay heat is presented. Numerical analyses to provide the sensitivity coefficients were made only for uniform changes of the parameters used for the estimations of fractional yields. The deviations of the decay heat from the reference were approximately proportional to the magnitudes of the parameter changes. Then, a general formula for the sensitivity coefficients was theoretically derived from the first order approximation and the correlation due to the charge conservation law between complementary fragments was taken into account. The sensitivity for the charge distribution width \( s_A \) was less than 1/10 of that for the most probable charge \( Z_p \). The evaluation of experimental errors for \( Z_p \) and \( s_A \) values were performed using radio-chemical data by Wahl and mass separator data by Clerc, and then the decay heat uncertainties were estimated by using the resultant \( Z_p \) and \( s_A \) experimental errors. Evaluated uncertainties of the decay heat for thermal neutron induced fission of \( ^{235}\text{U} \) after burst irradiation were 2.98\% at the cooling time of 2.7 s, 1.70\% at 8.2 s, 1.72\% at 68.7 s, 0.64\% at 700 s and 0.74\% at 12,000 s, respectively.

KEYWORDS: uranium 235, decay heat, sensitivity analysis, calculational errors, comparative evaluations, thermal neutrons, fission yields, charge distribution

I. INTRODUCTION

Precise prediction of the decay heat generated from fission products is a prerequisite to safe operation and design of the decay heat removal system equipped on power reactors, as well as the reactors themselves. Therefore much effort has been devoted to evaluation of the decay heat(1). However, it is necessary to evaluate the uncertainty of calculated decay heat using these nuclear data libraries since nuclear data such as decay constants, fission yields, or \( \beta \)-and \( \gamma \)-energies always have uncertainties. Schmitroth and Schenter(2) have evaluated the uncertainties due to the errors in nuclear data as a function of cooling time, based on a rather complicated, but rigorous method. Tasaka et al.(3) have developed a computer program HEAT5G for calculation of decay power and its uncertainty based on the ANS 5.1(4) decay heat standard. The program HEAT5G can take into account the \( ^{233}\text{U} \), \( ^{234}\text{U} \) and \( ^{239}\text{Pu} \) admixture contributions and it is a practical tool to estimate the decay power with uncertainty at an arbitrary operation period. However, detailed consideration on the uncertainties of basic parameters such as for charge distribution are out of their scope.

More than 1,000 fission products are con-
considered for the decay heat evaluation and they are classified into about 120 \( \beta \)-decay chains. The decay heat from an isotope is affected by all its precursors in the \( \beta \)-decay chain. The influences propagate from precursors to daughters through the \( \beta \)-decay chain calculation yielding atom densities as solutions, and they are implicit and non-linear functions of fission yields expressed in terms of mass yield and charge distribution. The isotopic decay energies are included as linear terms. Consequently, rigorous evaluation of the decay heat uncertainty is a complicated matter, requiring a huge amount of computation time.

The present work aims at giving a simplified method, instead of the rigorous one. The method is based on sensitivity coefficients. As an example, the uncertainty of decay heat in the thermal neutron induced fission of \( \text{U}^{235} \) is obtained as a function of cooling time after burst irradiation.

II. SIMPLIFIED METHOD FOR DECAY HEAT UNCERTAINTY EVALUATION

The decay heat due to fission products is given as a summation of isotopic contributions as

\[
H(t) = \sum_{i=1}^{I_{\text{max}}} H_i(t),
\]

\[
H_i(t) = (E \beta_i + E \gamma_i) \lambda_i N_i(t),
\]

where

\( H(t) \): Nuclear decay heat summed over all isotopes at cooling time \( t \) after a burst irradiation of neutrons

\( H_i(t) \): Decay heat of \( i \)-th isotope at time \( t \)

\( I_{\text{max}} \): Total number of isotopes

\( E \beta_i, E \gamma_i \): Average \( \beta \)- and \( \gamma \)-energies of \( i \)-th isotope

\( \lambda_i \): Decay constant

\( N_i(t) \): Atom density at cooling time \( t \).

The product \( \lambda_i N_i \) in the right-hand side of Eq. (1b) is the decay rate (activity) of the \( i \)-th fission product. An amount of the energy \( (E \beta_i + E \gamma_i) \) is released in a unit decay of the nucleus. For simplicity, the total energy release \( E_i \) is defined by

\[
E_i = E \beta_i + E \gamma_i.
\]

The atom density \( N_i \), as a function of mass yield, fractional yield, microscopic absorption rate and reactor power, is given as the result of the \( \beta \)-decay chain calculation. When the parameters are limited to the mass yield \( Y(A) \) of the fission products with mass number \( A \), the most probable charge \( Z_p \) and the charge distribution width \( \sigma_A \), the decay heat \( H(t) \) can be expressed by

\[
H = H[t, \{Y(A)\}, \{Z_p(A)\}, \{\sigma_A\}],
\]

where \( \{ \} \) is an abbreviation for the vector whose components indicate magnitudes of the physical parameters as a function of mass number \( A \). The deviation of the decay heat \( H \) due to changes of the vectors can be expressed by a Taylor expansion series as;

\[
\delta H = H[t, \{Y(A)\} + \delta Y(A)], \{Z_p(A) + \delta Z_p(A)\}, \{\sigma_A + \delta \sigma_A\}]

- \[H[t, \{Y(A)\}, \{Z_p(A)\}, \{\sigma_A\}]\] (4a)

\[
= \sum_{A=1}^{A_{\text{max}}} \sum_{n=1}^{N} \sum_{p,q,r} \frac{1}{n!} \left( \frac{\partial}{\partial \bar{W}} \right)^n \left( \frac{\partial}{\partial Y(A)} \right)^p \left( \frac{\partial}{\partial Z_p(A)} \right)^q \left( \frac{\partial}{\partial \sigma_A} \right)^r H,
\]

(4b)

\[
\Delta \bar{W} = \Delta W \cdot \frac{\partial}{\partial \bar{W}}, \quad (W = Y, Z_p, \sigma_A),
\]

where \( (\delta W \cdot \delta \bar{W})^t \) means \( s \) times operations of the derivative operator \( \delta W \cdot \delta \bar{W} \) to the function \( H \), and the summations with respect to \( p, q \) and \( r \) of Eq. (4b) are made for all possible combinations of integer values \( p, q \) and \( r \) under the condition \( p+q+r=n \), and \( A_{\text{max}} \) means the total number of mass chains. In practice, the second order approximation is acceptable and then the deviation \( \delta H \) can be shown by

\[
\frac{\partial}{\partial Y(A)} \left( \frac{\partial}{\partial Y(A)} \right) + \left( \frac{\partial}{\partial Z_p} \right) \left( \frac{\partial}{\partial Z_p} \right) + \left( \frac{\partial}{\partial \sigma_A} \right) \left( \frac{\partial}{\partial \sigma_A} \right) + \left( \frac{\partial}{\partial \bar{W}} \right) \left( \frac{\partial}{\partial \bar{W}} \right) \left( \frac{\partial}{\partial \bar{W}} \right)^t
\]

\[
= \sum_{A=1}^{A_{\text{max}}} \left[ \left( \frac{\partial}{\partial Y(A)} \right) \delta Y(A) + \left( \frac{\partial}{\partial Z_p} \right) \delta Z_p(A) \right] \]
where the second derivatives with respect to different variables in Eq. (6) mean mutual correlation terms. As a special case, if the magnitudes of the parameters are uniformly changed over all isotopes by $\varepsilon$ for $Y(A)$, $\mu$ for $Z_p$ and $\nu$ for $\sigma_A$, the decay heat change $\delta H$ becomes

$$
\delta H = \sum_{A=1}^{A_{\text{max}}} \left[ \varepsilon \left( \frac{\partial H}{\partial Y} \right) + \mu \left( \frac{\partial H}{\partial Z_p} \right) + \nu \left( \frac{\partial H}{\partial \sigma_A} \right) 
+ \varepsilon^2 \left( \frac{\partial^2 H}{\partial Y^2} \right) + \mu^2 \left( \frac{\partial^2 H}{\partial Z_p^2} \right) + \nu^2 \left( \frac{\partial^2 H}{\partial \sigma_A^2} \right) 
+ 2 \left( \varepsilon \mu \left( \frac{\partial H}{\partial Y \partial Z_p} \right) + \mu \nu \left( \frac{\partial H}{\partial Z_p \partial \sigma_A} \right) 
+ \nu \varepsilon \sum \left( \frac{\partial H}{\partial \sigma_A \partial Y} \right) \right] \right],
$$

(7)

In order to estimate the magnitude of the mutual correlation terms, the deviations of decay heats from a reference case were calculated by changing the $\mu$ and $\nu$ parameters by the computer program FPGR which can calculate the decay heat exactly by the "direct calculation method" using the JNDC nuclear data library whose fission yield data are based on the evaluated data by Rider and Meek(6) with minor corrections.

The resultant decay heats as a function of the most probable charge $Z_p$ and the charge distribution width $\sigma_A$ are shown in Fig. 1(a) and (b) respectively. These figures show that the decay heat is insensitive to the change of the $\sigma_A$-value in comparison with $Z_p$'s, i.e. less than 1/10 of the $Z_p$ sensitivity. Percentage deviations $\delta H/H$ of fission product decay heat for typical cooling times are shown in Fig. 2 as a function of the change of the most probable charge from the reference. The linearity of $\delta H/H$ vs. the relative change of the most probable charge $\delta Z_p/Z_p$ is good, and thus the change of decay heat $\delta H$ may be approximated by the first derivatives in Eq. (6) or (7).

In Eq. (6), the cross terms having different quantities such as $\delta Y \cdot \delta Z_p$ mean the correlation term between $\delta Y$ and $\delta Z_p$ errors. As
Szp, A and SsA, A are defined by which are functions of only mass number A for a chain with several isotopes. Consequently, Eq. (9) can be expressed in terms of “isotopic” sensitivity coefficient Szp, i as shown below:

\[
\delta H = \delta Y(A) \left( \frac{\partial H}{\partial Y(A)} \right) + \delta Zp(A) \left( \frac{\partial H}{\partial Zp(A)} \right) + \delta \sigma(A) \left( \frac{\partial H}{\partial \sigma(A)} \right),
\]

which are functions of only mass number A for a chain with several isotopes. Consequently, Eq. (9) can be expressed in terms of “isotopic” sensitivity coefficient Szp, i as shown below:

\[
\frac{\delta H}{H} = \sum_{A=1}^{A_{max}} S_{Y,A} \left( \frac{\partial H}{\partial Y(A)} \right) + \sum_{A=1}^{A_{max}} H_i \left( \frac{\partial H}{\partial Zp(A)} \right) + \sum_{A=1}^{A_{max}} S_{\sigma,A} \left( \frac{\partial H}{\partial \sigma(A)} \right),
\]

with

\[
S_{zp,i} = \frac{Zp,i}{H_i} \left( \frac{\partial H_i}{\partial Zp,i} \right),
\]

where the symbol i ∈ A means that all isotopic contributions belonging to the β-decay chain with the mass number A are summed up, and single summations for the first and third terms in Eq. (13) are based on the fact that Y(A) and σ(A) are defined by mass number A as a single value. The suffix i of the Zp, i is introduced for the simplicity of formulation although the Zp, i value is constant for a definite mass chain.

The chain sensitivity coefficients can be expressed by the isotopic ones as shown below

\[
S_{Y,A} = \sum_{i \in A} \frac{H_i}{H} S_{Y,i},
\]

\[
S_{zp,A} = \sum_{i \in A} \frac{H_i}{H} S_{zp,i},
\]

\[
S_{\sigma,A} = \sum_{i \in A} \frac{H_i}{H} S_{\sigma,i},
\]

where index i is used for chain yield Y and charge distribution width σ by extending its application to the chain quantities for sim-
plicity, i.e. in Eq. (15) \( S_{Y,i} \)'s are constants as long as only a definite mass chain is considered.

The sensitivity coefficient \( S_{x,A} \) (\( x = Y, Z_p \) or \( \sigma_A \)), as a function of \( A \) can be evaluated by relating the changes of the isotopic decay heats to the changes of atom densities \( N_i \) of \( i \)-th isotope. The sensitivity coefficient \( S_{x,A} \) based on the calculated decay heat for uniform changes of parameters can be obtained as shown below:

\[
S_{x,A} = \frac{x_A}{H} \sum_{i=1}^{I_{\text{max}}} E_i \lambda_i N_i(t),
\]

\[
= \frac{x_A}{H} \sum_{i=1}^{I_{\text{max}}} E_i \lambda_i N_i(t),
\]

\[
= \frac{x_A}{H} \sum_{i=1}^{I_{\text{max}}} E_i \lambda_i N_i(t) \frac{\partial}{\partial x_A} \ln[N_i(t)] ,
\]

\[
\approx \frac{x_A}{H} \sum_{i=1}^{I_{\text{max}}} H_i \frac{\ln[N_i(t, x_A + \delta x_A)] - \ln[N_i(t, x_A)]}{\delta x_A} ,
\]

(18a)

(18b)

(18c)

where \( x_A \) and \( \delta x_A \) stand for a generalized variable for \( Y(A), \sigma_A \) or \( Z_p \), and its small deviation, respectively. The partial derivative of Eq. (18c) is replaced by a finite difference of atom densities. This replacement is acceptable in the small range \( \delta x_A \) where linearity is good, and when the atom density is affected mainly by the variable \( \delta x_A \) of the fission fragment of interest. Therefore, explicit forms of the sensitivity coefficients are:

\[
S_{Z_{P,i}} = \frac{Z_{P,i}}{H \mu} \sum_{i=1}^{I_{\text{max}}} H_i \ln \left[ \frac{N_i(t, Z_{P,i} + \mu)}{N_i(t, Z_{P,i})} \right],
\]

\[
S_{\sigma_{A,i}} = \frac{\sigma_A}{H} \sum_{i=1}^{I_{\text{max}}} H_i \ln \left[ \frac{H(t, \sigma_A + \nu) - H(t, \sigma_A)}{H(t, \sigma_A)} \right],
\]

(19)

(20a)

(20b)

where \( I_{\text{max}} \) means the total number of isotopes considered as the sources of decay heat.

The sensitivity coefficient of \( \sigma_A \) to the isotopic decay heat, denoted by \( S_{\sigma_A,i} \), can be defined by

\[
S_{\sigma_A,i} = \frac{H_i}{H} S_{\sigma_A,i}.
\]

(20c)

In the following paragraphs, no consideration on the mass yield uncertainty is made since a reasonable method to correlate the mass yield with that of complementary fragments has not been found.

In the formulation of the sensitivity coefficients, the conservation law of nuclear charges has not been considered. The sum of the charges of complementary fragments and the charge of the emitted low mass particle is equal to that of the fissioning nucleus. This conservation law is still valid for the most probable charges of a pair of fission products as

\[
Z_P = Z_{P,i} + Z_{P,i}^* + z_L,
\]

\[
\approx Z_{P,i} + Z_{P,i}^*,
\]

(21a)

(21b)

where

- \( Z_P \): Nuclear charge of fissioning nucleus

- \( Z_{P,i}, Z_{P,i}^* \): Most probable charges of \( i \)-th fission fragment and its complementary one

- \( z_L \): Charge carried by low mass particles such as He in ternary fission.

Since the fission probability of ternary fission is about one-thousandth that of binary fission in lower energy fissions, the approximation of Eq. (21b), based on binary fission, is an accurate approximation.

Consequently, the changes of the most probable charges should satisfy the following equation as required by the charge conservation law:

\[
\delta Z_{P,i} + \delta Z_{P,i}^* = 0.
\]

(22)

Then the relative change of the most probable charge \( Z_{P,i}^* \) for the complementary fragment can be expressed by that of \( Z_{P,i} \) as

\[
\frac{\delta Z_{P,i}^*}{Z_{P,i}^*} = - Z_{P,i}^* \left( \frac{\delta Z_{P,i}}{Z_{P,i}} \right).
\]

(23)
With this correlation, the relative change of decay heat \((\delta H)_{zp}/H\) due to the change of the most probable charge \(Z_p\) can be expressed by an “effective” sensitivity coefficient \(S_{zp,i}\) combining the complementary fragments under the charge conservation law:

\[
\frac{(\delta H)_{zp}}{H} = \sum_{A=1}^{A_{\text{max}}} \left( \frac{H_i}{H} \right) \left[ S_{zp,i} \left( \frac{\delta Z_{p,i}}{Z_{p,i}} \right) + S_{zp,i}^* \left( \frac{\delta Z_{p,i}^*}{Z_{p,i}^*} \right) \right]
\]

where

\[
S_{zp,i} = \left( \frac{H_i}{H} \right) \left[ S_{zp,i} \frac{Z_{p,i}}{Z_{p,i}^*} \frac{H_i^*}{H_i} S_{zp,i}^* \right],
\]

\[
S_{zp,i} = \left( \frac{H_i}{H} \right) (1 - C_{A,i}) S_{zp,i}
\]

Therefore, the current problem to obtain the effective sensitivity coefficient is reduced to obtaining the isotopic sensitivity coefficient defined by Eq. (14) in an explicit form of the parameters \(Z_{p,i}\), \(\sigma_A\) and the others related to decay energy. The isotopic sensitivity coefficient can be expressed by the atom density \(N_i\) as shown below

\[
S_{zp,i} = \frac{\partial \ln |N_i(t)|}{\partial Z_{p}(A)}.
\]

The atom density \(N_i(t)\) is obtained from the \(\beta\)-decay chain equation:

\[
\frac{dN_i}{dt} = -(\lambda_i + \sigma_A \phi) N_i + (\lambda_{i-1} + \sigma_{ei_{i-1}} \phi) N_{i-1} + Y(A) f_i^{(c)} \langle \Sigma_f \phi \rangle,
\]

where \(Y(A)\): Mass yield as a function of mass number \(A\)

\(\langle \Sigma_f \phi \rangle\): Fission reaction rate

\(\lambda_i\): Decay constant of \(i\)-th fission product

\(f_i^{(c)}\): Fractional cumulative yield

\(\sigma_{ei_{i-1}}\): One group radiative capture cross section

\(t\): Cooling time.

As shown in APPENDIX, the solution of Eq. (28) under the steady-state condition is approximately given by

\[
N_i(t) = \frac{Y(A) \langle \Sigma_f \phi \rangle}{\lambda_i + \sigma_A \phi} f_i^{(c)},
\]

where fractional cumulative yield \(f_i^{(c)}\) is defined as the sum of fractional independent yields of \(\beta\)-decay precursors on a chain. Then, Eq. (27) can be expressed by

\[
S_{zp,i} = \frac{\partial \ln |f_i^{(c)}|}{\partial Z_{p,i}}.
\]

The empirical formula for fractional cumulative yield \(f_i^{(c)}\) can be expressed by the error function as

\[
f_i^{(c)} = \text{erf} \left( \frac{Z_i + 0.5 - Z_{p,i}}{\sqrt{2} \sigma_A} \right),
\]

with

\[
\text{erf}(x) = \frac{2}{\sqrt{\pi}} \int_0^x \exp(-t^2) dt
\]

and the \((n+1)\)-th derivative of the error function is a function of the Hermit function \(H_n(z)\) of the order \(n\) \((H_0=1)\):

\[
\frac{d^{n+1}}{dZ^{n+1}} \text{erf}(z) = (-1)^n H_n(Z) \exp(-Z^2),
\]

\((n=0, 1, 2, \ldots)\).

By substituting Eq. (31) into Eq. (30a) and differentiating the resultant equation with respect to \(Z_{p,i}\), the isotopic sensitivity coefficient \(S_{zp,i}\) can be expressed by

\[
S_{zp,i} = -(2/\pi)^{1/2} \frac{Z_{p,i}}{\sigma_A} \exp \left[ -\frac{\eta^2}{2} \right] / f_i^{(c)},
\]

with

\[
\eta = \frac{Z_i + 0.5 - Z_{p,i}}{\sqrt{2} \sigma_A}.
\]

The argument of the exponential function of Eq. (30b) results from the ratio of exponential functions as shown in Eq. (33) and by using the charge conservation law.

Therefore, by substituting Eq. (30b) into
Eq. (25), the effective sensitivity coefficient $S_{z_{p}, i}$ becomes a simpler form including only the ratio of isotopic decay heats and cumulative yields multiplied by an exponential function with respect to the chain length $\{Z_{i} - Z_{p}(A)\}$ as shown below:

$$S_{z_{p}, i} = \frac{H_{i}}{H_{i}} S_{z_{p}, i},$$  
(34a)

$$S_{z_{p}, i} = (1 - C_{A, i}) S_{z_{p}, i},$$  
(34b)

with the correction factor

$$C_{A, i} = \frac{H_{i}^{*}}{H_{i}} \frac{f_{i}^{(c)}}{f_{i}^{(c)*}} \exp \left( \frac{Z_{i} - Z_{p}(A)}{\sigma_{A}} \right),$$  
(35)

where $H_{i}$ and $H_{i}^{*}$ are the decay heats of the $i$-th isotope and its complement, and $f_{i}^{(c)}$ and $f_{i}^{(c)*}$ mean the cumulative yields.

Numerical examples of sensitivity coefficients are shown in Table 1 with the related quantities where the magnitudes of the uniform changes of parameters $\mu$ and $\nu$ are 0.11 and 0.03, respectively. In this table, nuclei having contributions to decay heat larger than 1% are shown. In Fig. 3 a comparison of the direct calculation and sensitivity methods are shown as a function of cooling time after burst irradiation of thermal neutrons to $^{235}$U. The reproducibility by the sensitivity method is good, except for the shorter cooling times where the discrepancy seems to be due to the approximation made for Eq. (30b). The ratios of the decay heat changes based on sensitivity method to direct calculation were 1.31, 1.20, 0.96, 1.33 and 0.54 for cooling times 2.7, 8.2, 68.7, 700 and 1,200 s, respectively.

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<th>$Z_{p} - Z_{i}$</th>
<th>$f_{i}^{(c)*}$</th>
<th>Complementary FP</th>
<th>$S_{z_{p}, i}$</th>
<th>$\frac{m}{\sigma_{A}}$</th>
<th>$S_{z_{p}, i}^{*}$</th>
<th>$\frac{m}{\sigma_{A}}$</th>
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<td>Ba145</td>
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(Note) $S_{z_{p}, i}$'s: Isotopic sensitivity coefficients are calculated by Eq. (30b).

$S_{z_{p}, i}$'s: Effective sensitivity coefficients based on Eq. (34b) taking into account the correlation between the most probable charges existing on the mass chains for complementary fragments under the condition of charge conservation law.
The errors of $Z_p$'s are assumed to be 0.11 for all fission products in a decay chain with a mass number $A$.

**Fig. 3** Comparison of decay-heat errors based on direct calculation and sensitivity methods for assumed error of most probable charge for assumed error of most probable charge $Z_p$.

In order to improve the sensitivity method, the sensitivity coefficients defined by Eq. (34b) for the uniform change of parameter are renormalized so that the sensitivity method gives the same magnitude as the direct calculation, as shown in Chap. III.

The sensitivity coefficients and related quantities for the decay heat at 2.7 s after the burst irradiation of thermal neutrons to $^{235}$U are shown in Table 1. The complementary nuclei in the table are estimated by taking into account the charge conservation law and the number of fission neutrons. The blank space found in the sixth column indicates there are no complementary nuclei in the table. The sensitivity coefficients for the heavy nucleus with a complementary one in the light mass region is not defined since the contribution of the complementary nucleus to the decay heat is considered in the light one by the correction factor $C_{A,t}$ applied to $S_{Z_{P,t}}$.

As shown in **Fig. 4**, the sensitivity coefficients $S_{Z_{P,t}}$ and $(1-C_{A,t})S_{Z_{P,t}}$ can be expressed by a smoothly varying function of the chain length $(Z-Z_p)$ as expected from the behavior of the analytical functions used for $S_{Z_{P,t}}$ of Eq. (30b) and $f^{(c)}$ defined in Eq. (31). The tendency of $S_{Z_{P,t}}$ to approach zero as the chain length increases means that the cumulative yield $f^{(c)}$ is saturating to unity and its derivative with respect to nuclear charge tends to zero. As evident from the relation between $C_{A,t}$ and $(1-C_{A,t})$, when the magnitude of the correction factor $C_{A,t}$ is greater than unity, the sign of the effective sensitivity coefficient $S_{Z_{P,t}}$ is inverted, because in such a case the contribution of the complementary nucleus to the decay heat is superior to that of the one of interest, and both contributions act so as to cancel each other as a result of the charge conservation law, as indicated by Eq. (22).

The sensitivity coefficient for $^{145}$Ba is far from the general trend in **Fig. 4**. This deviation is due to the fact that the sensitivity coefficient $S_{Z_{P,t}}$ is proportional to the $Z_p$-value when the nuclei have the same chain length $(Z-Z_p)$.

Total uncertainty of the decay heat relative to that of the reference case can be obtained by the error propagation law as

$$\frac{\delta H}{H} = \left\{ \sum_{A=1}^{A_{\text{max}}} \left[ \sum_{i=1}^{i_{\text{max}}} \left( S_{Z_{P,t}} \right)^2 \left( \frac{\delta Z_{P,t}}{Z_{P,t}} \right)^2 \right] \right\}^{1/2},$$

where the charge distribution width $\sigma_A$ is assumed to be constant for all isotopes in a chain with mass number $A$.

**III. NUMERICAL EXAMPLES OF DECAY HEAT UNCERTAINTY**

The most probable charge $Z_p$ is obtained from experimental data of fractional independent yield $f^{(i)}$ and/or cumulative $f^{(c)}$ yield compiled by Amiel and Feldstein. The errors of $Z_p$ are derived from the experimental errors of $f^{(i)}$ and/or $f^{(c)}$ by fitting the empirical formula,
Numerical data are shown in Table 1. The dash-dot line joining a circle to a box indicates these points are for complementary fragments.

Fig. 4 Sensitivity coefficients as functions of chain length

\[
f(Z) = \frac{1}{\sqrt{2\pi\sigma_A^2}} \exp \left(-\frac{(t-Z)^2}{2\sigma_A^2}\right) dt,
\]

where \( A \) and \( Z \) stand for the mass and atomic numbers of the fission product considered. While the \( Z_P \)-value is being searched for, the charge distribution width \( \sigma_A \) is initially assumed to be a constant value of 0.56(\( \pm \)0.06)\(^8\), and then the new charge distribution width \( \sigma_A \) is determined by using the resultant \( Z_P \)-value. A quantity \( \delta Z_P \) is introduced as an error of \( Z_P \), deviating from the value averaged over all isotopes belonging to the \( \beta \)-decay chain of interest. The distribution of errors denoted by \( \delta Z_P/Z_P \)'s relative to the \( Z_P \)-value is shown in Fig. 5. In Fig. 5(a), locations of each isotope are shown by the pair of mass and atomic numbers written in parentheses. Some duplicate combinations of \( (Z, A) \)'s are found since there are some experimental data with the same \( (Z, A) \) by different authors. Some data scattered from the center were not used for the fitting.

Figure 5(b) shows a normal distribution with the distribution width 0.456\% which is estimated from the frequency distribution shown in Fig. 5(a).

With the same procedure, an error distribution function is obtained for the charge distribution width \( \sigma_A^\text{ref} \) whose one standard deviation \( (\delta \sigma_A/\sigma_A)_{\text{std}} \) is 10.7\% on the basis of experimental data by Wahl\(^8\). The net changes of the most probable charge \( Z_P \), denoted by \( \delta Z_P \), and of the charge distribution width, denoted by \( \delta \sigma_A \), are obtained by

\[
\delta Z_P = \left( \frac{\delta Z_P}{Z_P} \right)_{\text{std}} Z_P^\text{ref},
\]

\[
\delta \sigma_A = \left( \frac{\delta \sigma_A}{\sigma_A} \right)_{\text{std}} \sigma_A^\text{ref},
\]

where \( Z_P^\text{ref} \) and \( (\delta Z_P/Z_P)_{\text{std}} \) mean the refer-
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</tr>
<tr>
<td>2.0</td>
<td>[ [ 93 ] [ 92 ] [ 193 ] [ 194 ] ]</td>
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<tr>
<td>1.5</td>
<td>[ [ 93 ] [ 92 ] [ 193 ] [ 194 ] ]</td>
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<tr>
<td>1.0</td>
<td>[ [ 94 ] [ 93 ] [ 194 ] [ 193 ] ]</td>
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<tr>
<td>0.8</td>
<td>[ [ 94 ] [ 93 ] [ 194 ] [ 193 ] ]</td>
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<tr>
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<td>[ [ 94 ] [ 93 ] [ 193 ] [ 194 ] ]</td>
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<tr>
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<td>[ [ 94 ] [ 93 ] [ 193 ] [ 194 ] ]</td>
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<tr>
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<td>[ [ 94 ] [ 93 ] [ 193 ] [ 194 ] ]</td>
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<td>[ [ 94 ] [ 93 ] [ 193 ] [ 194 ] ]</td>
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<tr>
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<td>[ [ 94 ] [ 93 ] [ 193 ] [ 194 ] ]</td>
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<td>[ [ 94 ] [ 93 ] [ 193 ] [ 194 ] ]</td>
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<tr>
<td>-1.5</td>
<td>[ [ 94 ] [ 93 ] [ 193 ] [ 194 ] ]</td>
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<td>-4.5</td>
<td>[ [ 93 ] ]</td>
</tr>
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</table>

The relative errors defined by $\delta Zr/Zr$'s are obtained by fitting an empirical formula to experimental, fractional independent and/or cumulative yields compiled by Amiel and Feldstein. [AA]: Mass number; [BB]: Atomic number.

Fig. 5(a) Relative error distribution of most probable charge for thermal neutron induced fission of $^{235}\text{U}$
ance \( Z_p \)-value and the standard deviation of the \( Z_p \)-errors as functions of mass number, respectively. \( \sigma_{A,C} \) and \( \langle \delta \sigma_{A,S} \rangle_{\text{std}} \) have analogous meanings to \( Z_p \)'s. In Fig. 6, the resultant \( \delta Z_p \) relative to \( Z_p^{\text{UCD}} \) (\( Z_p \)-value based on unchanged charge distribution postulate) is compared with the experimental data by Clerc et al.\(^{(9)}\) and Sigert et al.\(^{(10)}\) using the high resolution mass separator LOHENGRIN. Present results have more than twice as large experimental errors depending on the mass number as shown in Fig. 6. These larger values seem to be mainly due to using the radio-chemical data which give larger errors in comparison with the data by a mass spectrometer such as LOHENGRIN. For the numerical examples based on Eq. (36), the resultant \( Z_p \) standard deviation divided by a factor of 3 as an average discrepancy between the present and LOHENGRIN data is used to estimate the individual \( \langle \delta Z_{p,i} / Z_{p,i} \rangle \).

The decay heat sensitivity coefficients at cooling time 2.7 s after burst irradiation of

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**Fig. 5(b)** Normal distribution obtained by fitting probability density function to relative error distribution of most probable charge \( Z_p \).

**Fig. 6** Comparison of evaluated errors to those of experimental data by high resolution mass separator.
thermal neutrons to $^{239}$U are shown in Table 1. $S_{ZP,i}$ is the isotopic sensitivity coefficient based on Eq. (30b) and renormalized by the ratio of decay heat changes obtained by the sensitivity method to those by the direct calculation method due to a uniform change of parameter. The renormalization factor $R$ is defined by

$$R = \left[ \left( \frac{|DH_i^{(3)} - DH_i^{(1)}|}{DH_i^{(1)}} \right) / \sum_{i=1}^{N} \left( \frac{S_{ZP,i}}{Z_{P,i}} \right) \right]^{1/2},$$

where

- $DH_i^{(1)}$: Total decay heat based on direct calculation without change of $Z_P$-value
- $DH_i^{(2)}$: Total decay heat based on direct calculation with change of $Z_P$-value by 0.11.

Magnitudes of the renormalization factors are 0.93, 1.02, 0.61, 0.66 and 0.86 for cooling times 2.7, 8.2, 68.7, 700 and $1.20 \times 10^4$ s, respectively. $S_{ZP,i}'$ indicates the effective sensitivity coefficient based on Eq. (34b) for which the correlation between the most probable charge and its complementary one on a mass chain are considered under the condition of the charge conservation law. $C_{A,i}$ means the correlation factor.

Decay heat errors of individual isotope are evaluated from the error propagation law defined by Eq. (36). Decay heats and its uncertainties are shown in Table 2 at typical cooling times. “Reproducibility of Decay Heat” is provided to confirm the applicability of the sensitivity method in comparison with the direct calculation method. Uniform changes of 0.11 for $Z_P$ and 0.06 for $\sigma_A$ are applied to all isotopes in both methods and the resultant deviations of the decay heat from reference cases at each cooling time are estimated by summing over all isotopic deviations. As shown in Fig. 3, good agreement between both implies that the sensitivity method is

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<th>Cooling Time (s)</th>
<th>2.7</th>
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<th>68.7</th>
<th>700</th>
<th>12000</th>
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<td>1.34</td>
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<tr>
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<td>With Correlation</td>
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<td>1.71</td>
<td>1.27</td>
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<td>0.58</td>
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<tr>
<td>Without Correlation</td>
<td>2.61</td>
<td>2.06</td>
<td>2.11</td>
<td>0.88</td>
<td>1.01</td>
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(Notes) (a) Reproducibility of decay heat is examined by comparing the decay heat changes obtained by sensitivity and direct calculation methods, when the change of $-0.11$ for $Z_P$-value is used for both methods.
(b) Uncertainty of decay heat based on the sensitivity method by using Eq. (36) is for the actual error of $Z_P$'s obtained from the error distribution of experimental $Z_P$-values. Errors with and without correlations are estimated by using the sensitivity coefficients Eq. (34) with and without $C_{A,i}$, respectively. The correlation is defined between the most probable charges of the nucleus of interest and its complement under the charge conservation law.
(c) The uncertainties are given as a function of charge distribution width error, where $\delta \sigma_A=0.06$ is based on the experimental data by Wahl et al. and $\delta \sigma_A=0.03$ is used to indicate the measure of sensitivity of charge distribution width $\sigma_A$ to decay heat.
able to reproduce the change of decay heat for the uniform change of parameters such as $Z_P$ and $\sigma_A$.

In practice, the experimental errors of the most probable charge $Z_P$ and charge distribution width $\sigma_A$ occur at random and then the error propagation law Eq. (36) is recommended from a statistical viewpoint. The “Uncertainty of Decay Heat” based on Eq. (36) in Table 2 is for the $Z_P$ and $\sigma_A$ experimental errors. The uncertainty is a few percent and decreases with some oscillations, along with cooling time. The largest error occurs at cooling time 2.7 s.

Evaluated errors of decay heat by Schmitteroth and Schenter(2), reproduced from Fig. 4 of Ref. (2), are shown in Fig. 7(a) for $\delta \sigma = 0.06$ based on Wahl’s experiment and Fig. 7(b) for $\delta \sigma = 0.03$ as a function of cooling time in comparison with present result. The former case is recommended while the later one is shown as a measure of $\sigma$-sensitivity to the total uncertainty of decay heat. For instance, at $t=2.7$ s the present value is 2.98% while Schmitteroth and Schenter have 1.8%, but both get smaller with cooling time with some oscillations. These values are in general agreement although the present method is significantly simplified as compared to their method. It is noteworthy that the contribution of the error of the charge distribution width $\sigma_A$ for longer cooling time is small in comparison with $Z_P$'s and nearly constant in the range of the cooling time considered.

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Fig. 7(a), (b) Comparison of evaluated fission product decay heat uncertainties after burst irradiation of thermal neutrons to $^{235}$U
The correlation between the most probable charges of the complementary nuclei has been considered by introducing the effective sensitivity coefficient $S_{Z_p,i+}$, and the effects were shown in Table 2 as the uncertainty with or without correlation. According to the result based on the limited number of nuclei, the correlation effects in relative to the uncertainties without correlations were $-9.7\%$, $-17.5\%$, $-28.3\%$, $-30.4\%$ and $-33.3\%$ for cooling times $2.7$, $8.2$, $68.7$, $700$ and $1,200$ s, respectively.

IV. CONCLUSION

An error evaluation method on the sensitivity coefficients of decay heat against charge distribution parameters was developed. In this method, the sensitivity coefficients could be efficiently evaluated from the decay heat changes when charge distribution parameters were uniformly changed for all fission product.

Relative errors of the most probable charges obtained from the fractional independent and/or cumulative yields compiled by Amiel and Feldstein\(^{(5)}\) had a normal distribution and its one standard deviation $\langle \delta Z_p/Z_p \rangle_{\text{std}}$ was about $0.456\%$. The distribution of relative errors of charge distribution widths was obtained from the same experimental data whose one standard deviation $\langle \delta \sigma_A/\sigma_A \rangle_{\text{std}}$ was about $10.7\%$.

As a result of sensitivity analysis, the changes in decay heat were nearly proportional to the changes of the most probable charge or the change distribution width $\sigma_A$. The sensitivity to $\sigma_A$ was less than $1/10$ of that of the most probable charges.

Evaluated uncertainties of the decay heat for thermal neutron induced fission of $^{235}\text{U}$ after the burst irradiation were $2.98\%$ at $2.7$ s, $1.70\%$ at $8.2$ s, $1.72\%$ at $68.7$ s, $0.64\%$ at $700$ s and $0.78\%$ at $12,000$ s, respectively, when charge was conserved between the fission fragments of interest and its complementary one.

The present work aimed at a calculational method to evaluate the uncertainty of calculated decay heat due to the experimental errors of basic parameters for charge distribution. In this work, no consideration on the mass yield uncertainty as well as decay constants, energy releases by $\beta$- and $\gamma$-rays were made. Actual uncertainty of decay heat should be evaluated by taking into account of all these sources of errors.

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[APPENDIX]

The decay chain equation if the i-th isotope with the atom density \( N_i(t) \) is expressed by

\[
\frac{dN_i(t)}{dt} = -\lambda_{ai}N_i(t) + \lambda_{ei-1}N_{i-1}(t) + Y(A)\langle \Sigma_f \phi \rangle f_t^{(i)},
\]

where the effective decay constants denoted by \( \lambda_{ai}^*, \lambda_{ei-1}^* \) and other quantities are defined by

\[
\lambda_{ai}^* = \lambda_i + \sigma_{ai}\phi,
\]
\[
\lambda_{ei-1}^* = \lambda_{e,i-1} + \sigma_{ei-1}\phi,
\]

where \( \lambda_i \): Decay constant of i-th isotope
\( \sigma_{ai} \): Microscopic neutron absorption cross section
\( \sigma_{ei-1} \): Microscopic radiative capture cross section
\( N_i(t) \): Atom density of i-th isotope at time \( t \)
\( Y(A) \): Mass (chain) yield
\( \langle \Sigma_f \phi \rangle \): Fission reaction rate
\( f_t^{(i)} \): Fractional independent yield
\( \phi \): Neutron flux.

When the system is in the steady state, the atom density \( N_i(t) \) can be obtained from Eq. (A1) as

\[
N_i(t) = N_0 \frac{\lambda_{ei}^*}{\lambda_{ei}^* + \lambda_{eh}^*} \prod_{k=1}^{l} \left( \frac{\lambda_{ek}^*}{\lambda_{eh}^*} \right)
\]

\[+ \frac{Y(A)\langle \Sigma_f \phi \rangle}{\lambda_{ei}^*} \prod_{n=1}^{l} f_t^{(i)} \left( \frac{\lambda_{ek}^*}{\lambda_{eh}^*} \right).\]

(A2)

As a special case, if the neutron absorption rate is assumed to be small, i.e. \( \lambda_{ai}^* = \lambda_{ei}^* \), the atom density \( N_i(t) \) can be expressed by a simpler form as

\[
N_i(t) = N_0 + \frac{Y(A)}{\lambda_{ai}^*} \langle \Sigma_f \phi \rangle \prod_{n=1}^{l} f_s^{(i)},
\]

(A3)

where the atom density \( N_0 \) and the fractional independent yield \( f_s^{(i)} \) for the first isotope on the linear decay chain can be chosen to be zero such that its fission yield is negligibly small, since its atomic number is very far from the most probable charge. Consequently, The atom density \( N_i(t) \) in the steady state can be approximated by

\[
N_i(t) \approx \frac{Y(A)}{\lambda_{ai}^*} \langle \Sigma_f \phi \rangle f_s^{(i)},
\]

(A4)

where fractional cumulative yield \( f_s^{(i)} \) is defined by the summation of the fractional independent yields as shown by Eq. (A3).