Prediction of the Total Decay Heat from Fast Neutron Fission of $^{235}$U and $^{239}$Pu


**Abstract**—The analytical prediction of the decay heat results from the fast neutron fission of actinides was initiated under a project, 10-MAT1134-3, funded by king Abdulaziz City of Science and Technology (KASCT), Long-Term Comprehensive National Plan for Science, Technology and Innovations, managed by a team from King Abdulaziz University (KAU), Saudi Arabia, and supervised by Argonne National Laboratory (ANL) has collaborated with KAU’s team to assist in the computational analysis. In this paper, the numerical solution of coupled linear differential equations that describe the decays and buildsups of minor fission product actinides, MFA, has been used to predict the total decay heat and its components from the fast neutron fission of $^{235}$U and $^{239}$Pu. The reliability of the present approach is illustrated via systematic comparisons with the measurements reported by the University of Tokyo, in YAYOI reactor.

**Keywords**—Decay heat, fast neutron fission, and Numerical Solution of Linear Differential Equations.

I. INTRODUCTION

Accurate prediction of total decay heat and its time dependence is required in studies of loss of coolant accidents, and also in connection with the transportation and storage of spent fuel. At short cooling times, the main component is due to the decay of the fission products and also to $^{235}$U and $^{239}$Np decay, which are important for cooling times up to 15 days.

Many of the neutron-rich nuclei in question have also considerable current spectroscopic and astrophysical importance. In nuclear structure, a key issue of interest is the modification of shell structure in nuclides with large neutron excesses. In astrophysics, the challenge is to more clearly define the rate and trajectory of r-process nucleosynthesis in order to better delineate possible sites for this type of element production. Whatever the motivation, improved $\beta$-decay spectroscopy of neutron-rich fission fragments is needed.

During the last 20 years enormous progress has been made in nuclear spectroscopy, especially in improving the sensitivity for “in-beam” gamma ray spectroscopy [1, 2]. The production cross-sections required for detailed spectroscopy have fallen from millibarns to tens of nanobarns, an improvement of five orders of magnitude. With a few notable exceptions these advances have not been applied to $\beta$-decay spectroscopy studies. Some aspects of the new large gamma arrays, like Gammasphere, are not required for decay spectroscopy, (for example their very high angular granularity used for Doppler correction). Other aspects, like their high coincidence efficiency, and capability for total energy calorimetry, can be critical in resolving complicated decay patterns for nuclei far from the line of stability whose decay Q-values are significantly large. Combining data from modern multi-detector germanium arrays with high quality calorimetry from Total Absorption Gamma Spectrometers (TAGS) has already proven (in a few interesting cases) [3, 4] to be very powerful and complimentary in revealing both the gross decay features, like the total decay strength functions, and the details of the daughter nuclear structure. It is this path of research it is proposed to pursue at Argonne, using a TAG spectrometer for calorimetry.

The experimental data are particularly scarce for short cooling times (less than 3000 s) where the decay of neutron-rich FP dominates owing to the large $\beta$-decay Q values (~4-10 MeV) and the fact that $\beta$-decay feeding intensities into the high-energy region of the daughter nuclei are frequently missing (“pandemonium effect”) [5]. This is the case for almost half of all known FP involved in the fission process (~1200 nuclides). These missing intensities account for about 20-40 % loss in energy releases. In order to compensate for such a loss, due to paucity of experimental data, a gross beta-decay modeling is frequently used in the decay data libraries [6, 7], and therefore these libraries become “contaminated” by theoretical predictions rather than been on a firm experimental footing. Attempts made in past to resolve data deficiencies using high-resolution $\gamma$-ray spectroscopy techniques were only partly successful, due to the low efficiency and sensitivity of the detector systems used in these measurements and the lack of pure, and intense sources.

In the present work, following our approach that has been successfully used before in predicting the decay heat from Cm isotopes in the mixed oxide nuclear fuel, MOX, we continue reporting the data obtained in our project from the fast fission of $^{235}$U and $^{239}$Pu [8].

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II. METHODOLOGY

The algorithm of the present approach is divided into three steps. In the first step, the building the branching ratio, the decay constant and the independent fission yield matrices for the isotopes in a nuclear fuel can be calculated as; 

\[
\Delta = \begin{bmatrix}
-\lambda_1 & \lambda_2 b_{2,1} & \cdots & \lambda_m b_{m,1} \\
\vdots & \ddots & \ddots & \vdots \\
\lambda_1 b_{1,1} & \cdots & \lambda_m b_{m,1} & \vdots \\
\vdots & \ddots & \ddots & \vdots \\
\lambda_1 b_{1,m} & \cdots & \lambda_m b_{m,m} & -\lambda_m
\end{bmatrix}
\]  

(1)

Where, \( b_{j,i} \) is the branching ratios matrix to nuclide \( i \) per decay of nuclide \( j \), which satisfied that \( 0 \leq b_{j,i} \leq 1 \) and \( b_{j,i} = 0 \), \( \lambda_i \) is the decay constant of the \( i \)-th nuclide, and the independent fission yield \( Y_i \) of nuclide \( i \) at the initial condition is

\[
Y_i = \begin{bmatrix}
Y_{i,1} \\
Y_{i,2} \\
\vdots \\
Y_{i,m}
\end{bmatrix}
\]

(2)

In the second, we feed the matrices from Eq. 1 and 2 into our inventory code which has been written in MATLAB to calculate the number of nuclides after cooling time, \( N_i(t) \), by solving the Bateman ordinary differential equations systems of initial value problem in the form;

\[
\frac{d}{dt}N_i(t) = -\lambda_i N_i(t) + \sum_{j=2, j \neq i}^{m} b_{j,i} \lambda_j N_j(t) + Y_i
\]

(3)

Whereas, in the last step, the decay heat power in (MeV/fission/s) following time \( t \) after a fission burst of curium isotopes in a nuclear fuel can be calculated as;

\[
f(t) = \sum_{i} \bar{E}_{\beta,i} \lambda_i N_i(t)
\]

(4)

where, \( \bar{E}_{\beta,i} = \left( E_{\beta,i} + E_{\gamma,i} + E_{\alpha,i} \right) \), \( E_{\beta,i} \), \( E_{\gamma,i} \) and \( E_{\alpha,i} \) are the average \( \beta \)-, \( \gamma \)- and heavy particles energies per disintegration of nuclide \( i \). Those two average values were calculated from the decay schemes by the following relations; \( E_{\gamma,i} = \sum_i E_i I_i \) and \( E_{\beta,i} = \sum_i I_i \langle E_{\beta} \rangle_i \), where \( E_i \) is the gamma ray energy, \( I_i \) is the corresponding gamma ray emission probability, and \( \langle E_{\beta} \rangle_i \) is the mean energy of the \( \beta \) continuum populating level \( i \). Input data required to calculate the FP decay heat are the independent fission-yield, \( Y_i \) and decay data (branching ratios, \( b_{j,i} \), decay constants, \( \lambda_i \), and average \( \beta \)- and \( \gamma \)-decay energies releases, \( E_{\beta,i} \), \( E_{\gamma,i} \) which are extracted from ENDF/B-VII.I.

The number of nuclides \( N(t) \) after shutting down the reactor can be evaluated by solving Eq. 3 numerically using the fourth order Runge-Kutta method, RKM, for initial value problem of the ordinary differential equations of the decay system, \( N'_i(t) = f(t, N_i(t)) \), [9, 10]. In this method, a function \( f(t, N_i(t)) \) is evaluated several times for different time steps, \( dt \), between \( t_m \) and \( t_{m+1} \), and values of \( N(t) \) obtained by adding linear combinations of the values of \( f \) to \( N_m \). MATLAB program has the ordinary differential equation solver capabilities of the form "ode45" with digits nn indicating the order of the underlying method. We have used "ode23" solver capability, indicating that two simultaneous single-step formulas, one of second order and one of third order, are involved. More information about the present approach is in [11, 12].

III. RESULTS AND DISCUSSION

The total decay heat results from the fast neutron fission of \(^{235}\)U and \(^{239}\)Pu has been calculated using the present algorithm, using the decay data and fission yield data from ENDF/B-VII.I. University of Tokyo is shown in Figs. 1 and 2, respectively, [13].

The discrepancies from the reported measured values calculated by the following Eq. are shown in Figs. 3 and 4, for the two fissile nuclides, respectively.

\[
\Delta{\%} = \frac{f(t) \mid_{\text{measured}} - f(t) \mid_{\text{predicted}}}{f(t) \mid_{\text{measured}}}
\]

(5)

The highest calculated discrepancy values were less than 8.5 % and 10 % for \(^{235}\)U- and \(^{239}\)Pu- fast induced fission, respectively.

The contribution of each nuclide to the total FP decay heat following time after fission process for nuclides which contribution is larger than 1% is calculated at different cooling time 10 s and 1000 s, for \(^{235}\)U and \(^{239}\)Pu and presented in Table (1 and 2). The tabulated results show that the highest contributors for \(^{235}\)U- fast and \(^{239}\)Pu- fast are \(^{108}\)Nb and \(^{95}\)Y, respectively. Whereas, the least
contributors are \((^{132}\text{m} Sb\) and \(^{105}\text{Tc}\)) and \((^{106}\text{Nb}\) and \(^{95}\text{Y}\)), respectively.

By comparing the decay data of the tabulated results from the ENDF data sheets, it shows also some nuclides that might be suffering from the "pandemonium effect", which are highlighted in the tables.

IV. CONCLUSION

The numerical evaluation of the number of nuclides after a cooling time \(i\) and the combination between the hybrid and summation methods offers a good methodology to calculate the total decay heat (MeV/fission/s) produced by the curium isotopes in the mixed oxide nuclear fuel using the decay data and fission yield from ENDF/B-VII.1 (2011) database. Moreover, the listed nuclides which have the greatest contribution to the decay heat can be useful for deciding which nuclides need precise measurements as the highlighted ones in Table I.
<table>
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<th>Nuclide</th>
<th>Contribution %</th>
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REFERENCES


