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

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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 buildups 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. Introdution

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 *β-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 *β-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 β^--decay Q values (~4-10 MeV) and the fact that β^--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 γ–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 under consideration from the ENDF/B-VII.I database by applying the matrix form as follow;

$$\Delta = \begin{bmatrix} -\lambda_1 & \lambda_2 b_{2,1} & \cdots\cdots & \lambda_m b_{m,1} \\ \vdots & \cdots & \cdots & \cdots \\ \lambda_1 b_{1,i} & \cdots & \cdots & \lambda_m b_{m,i} \\ \vdots & \cdots & \cdots & \cdots \\ \lambda_1 b_{1,m} & \cdots & \cdots & -\lambda_m \end{bmatrix} \quad (1)$$

Where, $b_{j,i}$ is the is the branching ratios matrix to nuclide i per decay of nuclide j, which satisfied theat $0 \le b_{j,i} \le 1$ and $b_{i,i} = 0$, λ_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

$$Y_i = \begin{bmatrix} Y_1 \\ Y_2 \\ \vdots \\ \vdots \\ Y_m \end{bmatrix} \quad (2)$$

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;

$$\frac{d}{dt} N_i(t) = -\lambda_i N_i(t) + \sum_{j=1,\, j\neq i}^{m} b_{j,i} \lambda_j N_j(t) + Y_i \quad (3)$$

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;

$$f(t) = \sum_i \overline{E}_i^{\beta,\gamma,\alpha} \lambda_i N_i(t) \quad (4)$$

where, $\overline{E}_i^{\beta,\gamma,\alpha} = \left(\overline{E}_{\beta,i} + \overline{E}_{\gamma,i} + \overline{E}_{\alpha,i}\right)$, $\overline{E}_{\beta,i}$, $\overline{E}_{\gamma,i}$ and $\overline{E}_{\alpha,i}$ are the average β-, γ- and heavy particles energies per disintegration of nuclide i. Those two average values were calculated from the decay schemes by the following relations; $\overline{E}_{\gamma,i} = \sum_i E_i I_i$ and $\overline{E}_{\beta,i} = \sum_i I_i \langle E_\beta \rangle_i$, 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 β 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, λ_i, and average β- and γ- decay energies releases, $\overline{E}_{\beta,i}$, $\overline{E}_{\gamma,i}$) 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, Δt, 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 "odenn" 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.

$$\Delta\% = \frac{f(t)\,|_{PST} - f(t)\,|_{measured}}{f(t)\,|_{measured}} \quad (5)$$

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 (^{100}Nb and ^{95}Y) and (^{100}Nb and ^{95}Y), respectively. Whereas, the least

contributors are (^{132m1}Sb and ^{105}Tc) and (^{100}Nb and ^{95}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.

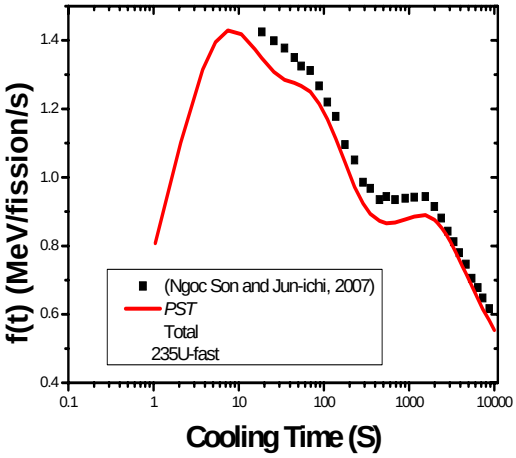

Fig. 1 Total- decay heat for ^{235}U- fast induced fission

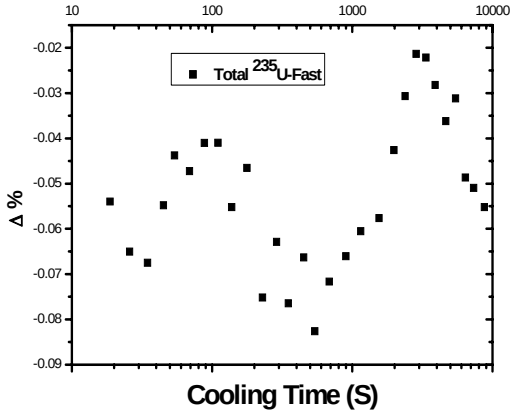

Fig. 2 Discrepancies Δ % between the calculated total- decay heat for ^{235}U- fast induced fission and the measured ones in YAYOI (Ngoc Son and Jun-ichi, 2007)

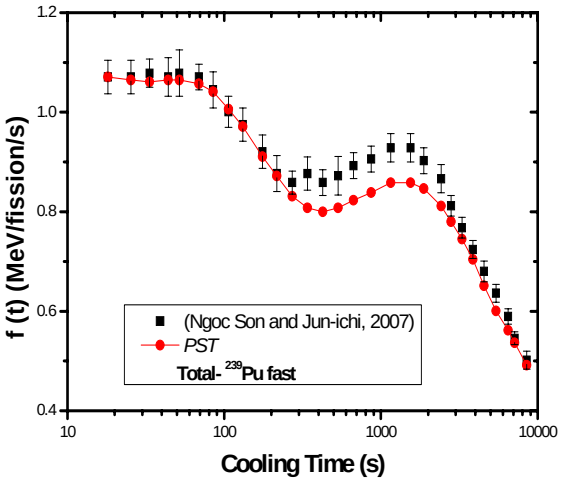

Fig. 3 Total- decay heat for ^{239}Pu- fast induced fission

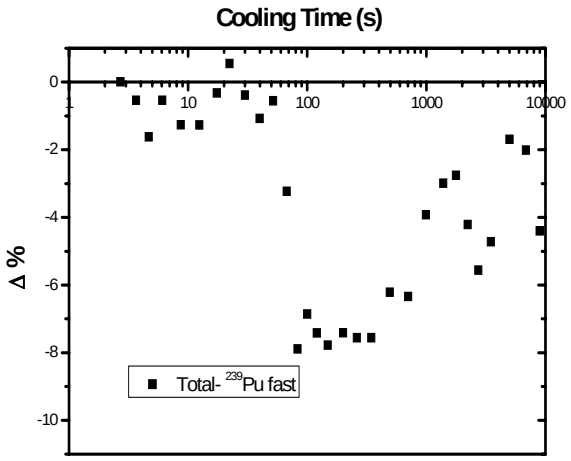

Fig. 4 Discrepancies Δ % between the calculated total- decay heat for ^{239}Pu- fast induced fission and the measured ones in YAYOI (Ngoc Son and Jun-ichi, 2007)

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 I
MAIN CONTRIBUTIONS TO THE TOTAL DECAY HEAT FROM ^{235}U- AND ^{239}PU- FAST INDUCED FISSION CALCULATED AT DIFFERENT COOLING TIMES BY THE PRESENT APPROACH

^{235}U-fast				^{239}Pu-fast			
10 S		1000 S		10 S		1000 S	
Nuclide	Contribution %	Nuclide	Contribution %	Nuclide	Contribution %	Nuclide	Contribution %
100 Nb	6.017	95 Y	7.163	100 Nb	6.675	104 Tc	9.561
92 Rb	5.274	93 Sr	6.785	101 Nb	5.325	102 Tc	5.934
96 Y	4.762	89 Rb	6.695	96 M1 Y	3.256	101 Mo	5.608
93 Rb	4.585	94 Y	6.561	92 Rb	3.215	95 Y	5.258
101 Nb	3.537	139 Cs	5.300	96 Y	2.958	139 Cs	4.619
97 Y	3.452	138 Cs	4.610	99 Nb	2.729	138 Cs	4.578
95 Sr	2.946	138 Xe	4.359	97 Y	2.613	94 Y	4.541
91 Kr	2.945	141 Ba	4.211	93 Rb	2.588	105 Tc	4.291
102 Nb	2.695	101 Mo	4.162	100 Zr	2.578	93 Sr	4.255
143 Ba	2.614	143 La	3.903	102 M1 Nb	2.509	141 Ba	3.725
100 Zr	2.359	133 Te	3.762	143 Ba	2.501	138 Xe	3.542
88 Br	2.207	102 Tc	3.734	95 Sr	2.413	143 La	3.055
99 Nb	2.189	142 Ba	3.288	102 Nb	2.174	142 Ba	2.760
141 Cs	2.098	104 Tc	2.920	98 Nb	2.081	89 Rb	2.701
146 La	2.022	131 Sb	2.215	106 Tc	2.008	134 I	2.429
145 Ba	1.898	137 Xe	1.955	141 Cs	2.002	133 Te	2.376
90 Kr	1.646	134 I	1.822	106 Mo	1.599	131 Sb	2.258
98 Nb	1.588	134 Te	1.695	138 I	1.506	101 Tc	1.868
94 Rb	1.587	146 Pr	1.653	103 Mo	1.470	137 Xe	1.838
140 Xe	1.571	90 Rb	1.540	108 Tc	1.468	146 Pr	1.467
96 M1Y	1.569	147 Pr	1.528	135 Te	1.462	147 Pr	1.463
138 I	1.488	142 La	1.526	105 Mo	1.412	142 La	1.400
145 La	1.469	101 Tc	1.372	145 La	1.408	134 Te	1.288
144 La	1.461	89 Kr	1.360	144 La	1.371	133 M1 Te	1.178
89 Br	1.369	132 M1 Sb	1.073	107 Tc	1.360	102 Mo	1.072
144 Ba	1.364	105 Tc	0.969	140 Xe	1.292	108 Rh	1.047
135 Te	1.354	84 Br	0.825	91 Kr	1.252	130 Sb	0.999
139 Xe	1.208	87 Kr	0.811	140 Cs	1.209	107 Rh	0.997
137 I	1.141	130 Sb	0.799	139 Xe	1.207	107 Ru	0.799
91 Rb	1.139	133 M1 Te	0.737	137 I	1.204	130 M1 Sb	0.789
140 Cs	1.074	90 M1 Rb	0.707	104 Nb	1.179	131 Te	0.655
101 Zr	1.072	92 Sr	0.694	144 Ba	1.155	132 M1 Sb	0.642
140 Cs	1.072	146 Ce	0.682	146 La	1.043	146 Ce	0.585
101 Zr	1.072	92 Sr	0.694	101 Zr	1.026	139 Ba	0.580
		146 Ce	0.682			152 Pm	0.544

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REFERENCES

[1] P. Reiter, T. L. Khoo, I. Ahmad, A. V. Afanasjev, A. Heinz, T. Lauritsen, C. J. Lister, D. Seweryniak, P. Bhattacharyya, P. A. Butler, M. P. Carpenter, A. J. Chewter, J. A. Cizewski, C. N. Davids, J. P. Greene, P. T. Greenlees, K. Helariutta, R.-D. Herzberg, R. V. F. Janssens, G. D. Jones, R. Julin, H. Kankaanpää, H. Kettunen, F. G. Kondev, P. Kuusiniemi, M. Leino, S. Siem, A. A. Sonzogni, J. Uusitalo, and I. Wiedenhöver, " Structure of the Odd-*A*, Shell-Stabilized Nucleus $^{253}_{102}$No," Phys. Rev. Lett. Vol. 95, pp. 032501/1-4, 2005.

[2] D. Seweryniak, N. Davids, C. Robinson, J. Woods, B. Blank et al, "Particle-core coupling in the transitional proton emitters $^{145, 146, 147}$Tm," Eur. Phys. J, vol. A25, Supplement 1, pp. 159, 2005. C. L. Duke, P. G. Hansen, O. B. Nielsen, G. Rudstam, "Strength-function phenomena in electron-capture beta decay," ISOLDE COLLABORATION, CERN Geneva, Switzerland,. Nucl. Phys., vol. A151, pp. 609, 1970.

[3] R. C. Greenwood, R.G. Helmer, M. H. Putnam, K. D. Watts, "Measurement of β^--decay intensity distributions of several fission-product isotopes using a total absorption γ-ray spectrometer," Nucl. Instr. and Meth. vol. A390, pp. 95. 1997.

[4] J. A. Hardy, L. C. Carraz, B. Jonson, P. G. Hansen, P.G., "The essential decay of pandemonium: A demonstration of errors in complex beta-decay schemes," Phys. Lett., vol. B71, pp. 307, 1977.

[5] T. Yoshida, R. Najasima, *J. Nucl. Sci. and Technol*., vol 18, pp. 393, 1981.

[6] O. Masahico, K. Shin-Chi, M. Katsufomi, N. Takashi, M. Toshiaki, "Analysis of Curium Isotopes in Mixed Oxide Fuel Irradiated in Fast Reactor," J. Nucl. Sci. Tech., vol. 38, pp. 912, 2001.

[7] S. Nafee, A. Al-Ramady and S. Shaheen, "Decay Heat Contribution Analyses of Curium Isotopes in the Mixed Oxide Nuclear Fuel," World Academy of Science, Engineering and Technology J., vol 68, pp. 2238, 2012.

[8] L. F. Shampine, "*Numerical Solution of Ordinary Differential Equations,*" Chapman and Hall, New York, 1994.

[9] L. F. Shampine and M. W. Reichelt, *Journal on Scientific Computing*, vol 18, 1997, pp. 1.

[10] K. E. Brenan, S. L. Campbell, and L. R. Petzold, "*Numerical Solution of Initial Value Problems in Differential-Algebraic Equations*," SIAM, Philadelphia, 1996.

[11] P. Bogacki and L. F. Shampine, "A 3(2) pair of Runge - Kutta formulas,"*Applied Mathematics Letters*, vol 2, 1989.

[12] Ngoc Son, P., and Jun-ichi, K., 2007. Application Program for Fission Product Decay Heat Calculations, JAEA-Data/ Code.