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Decay Heat Contribution Analyses of Curium Isotopes in the Mixed Oxide Nuclear Fuel

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Abstract**—**The mixed oxide nuclear fuel (MOX) of U and Pu contains several percent of fission products and minor actinides, such as neptunium, americium and curium. It is important to determine accurately the decay heat from Curium isotopes as they contribute significantly in the MOX fuel. This heat generation can cause samples to melt very quickly if excessive quantities of curium are present. In the present paper, we introduce a new approach that can predict the decay heat from curium isotopes. This work is a part of the project funded by King Abdulaziz City of Science and Technology (KASCT), *Long-Term Comprehensive National Plan for Science, Technology and Innovations*, and take place in King Abdulaziz University (KAU), Saudi Arabia. The approach is based on the numerical solution of coupled linear differential equations that describe decays and buildups of many nuclides to calculate the decay heat produced after shutdown. Results show the consistency and reliability of the approach applied.

Keywords**—**Decay heat, Mixed oxide nuclear fuel, Numerical Solution of Linear Differential Equations, and Curium isotopes

I. Introduction

RELIABLE quantification of the decay heat induced by nuclear fission is a critical factor in the design of nuclear facilities for electricity generation and for the post-irradiation handling of nuclear fuels (fuel discharge, storage, transport, and reprocessing, and waste handling). The total decay heat as a function of cooling time impacts both the safe operation and various legislative, and economic aspects of nuclear power generation [1-3]. New reactor design studies have highlighted the need for high quality experimental decay data, and careful propagation of uncertainties associated with these measurements. New reactor fuels under consideration demand a broader range of decay data than was previously required. These new reactor studies require decay data for many nuclides (primarily fission products and actinides), which include half-lives, mean beta and gamma-ray energies, feeding intensities, delayed neutrons, and sound estimates of their uncertainties [1, 4-6].

In nuclear energy applications, the evaluation of the decay heat have been usually performed using the summation (microscopic) method, where one follows the buildup and decay of about thousand fission products (FP) from the known fission yield and decay data [1].

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In this approach the decay heat (including both the beta- and gamma-ray components) is calculated from the known FP decay schemes as the total sum of the energy releases from all individual β^--decaying nuclides existing at a relevant cooling time. Although extensive measurement efforts have been developed over the past fifty years in response to the increasing need of the applied nuclear reactor community for accurate decay data, differences still exist between the decay heat standards and experiments, as well as between different decay data libraries [2, 3, 6, 7]. 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") [8]. 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 [7, 9], 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.

The Total Absorption Gamma-ray Spectrometry (TAGS) method [10, 11], which is in principle free from the "pandemonium" problem, has been applied in the past only to about 50 cases [10], albeit this technique is compromised when isomers are presented or β^--decay delayed neutrons are emitted. The urgent need for improved decay data on neutron-rich FP for Advanced Fuel Cycles applications has been recently outlined at IAEA consultants' meetings on "Beta-decay and Decay Heat" [2, 3]. The low irradiated mixed oxide fuel (MOX) of U and Pu usually contains several percent of fission products (FP) and minor actinides (MA) such as neptunium (Np), americium (Am) and curium (Cm). Curium is considered a key element in the multi - recycling of nuclear fuel because its seven isotopes have high radiotoxity that makes the nuclear waste disposal more difficult.

The curium isotopes are divided into lower mass (^{242}Cm, ^{243}Cm and ^{244}Cm) and higher mass (^{245}Cm, ^{246}Cm and ^{247}Cm) groups [12]. ^{244}Cm and ^{245}Cm are the most abundant isotopes MOX fuel of concentrations varies significantly with the fuel burnup [13]. Therefore, we are mainly concerns about the accurate determination of the decay heat of ^{244}Cm and ^{245}Cm and their contributions to the heat produced in the nuclear fuel within the first 10^5 seconds after shutdown.

In the present work, we proposed a new technique which uses three steps to evaluate the decay heat function. In the first step, we build the branching ratio, the decay constant and the independent fission yield matrices for the isotopes under consideration from the ENDF/ B-VII.I database. The second step concerning feeding those matrices into our inventory code to calculate the number of nuclides after cooling time by solving the ordinary differential equations of initial value problem. Whereas, in the last step we used the ordinary summation method to calculate the decay heat.

II. Methodology

The most common solution for the differential equations that describe the radioactive decay following the fission in a nuclear power reactor is known as the Bateman solution. Bateman used Laplace transforms to solve these differential equations' system [14].

$$\frac{d}{dt}N_i(t) = -\lambda_i N_i(t) + P_i + Y_i \tag{1}$$

Where, P_i is the buildups of the nuclides and is given by $P_i = \sum_{j=1,\, j\neq i}^{m} b_{j,i}\lambda_j N_j(t)$, λ_i is the decay constant of the *i*-th nuclide, $N_i(t)$ is the number of nuclide *i* at cooling time t and Y_i is the independent fission yield of nuclide *i*. $b_{j,i}$ is the is the branching ratios matrix to nuclide *i* per decay of nuclide *j*, which satisfied the following conditions;

$$0 \le b_{j,i} \le 1 \text{ and } b_{i,i} = 0 \tag{2}$$

Also, if the chain under consideration contains all daughters of nuclide *i*, then;

$$\sum_j b_{j,i} = 1 \tag{3}$$

Generally, $b_{j,i} \neq b_{i,j}$ unless $b_{j,i} = b_{i,j} = 0$.

By applying the matrix form, equation (1) can be rewritten in the form [15];

$$\frac{d}{dt}N_i(t) = N_i' = \Delta N_i(t) \tag{4}$$

Where,

$$N_i(t) = \left[N_1(t) N_2(t) \cdots N_i(t) \cdots N_m(t)\right]^T \tag{5}$$

$$\text{And } \Delta = \begin{bmatrix} -\lambda_1 & \lambda_2 b_{2,1} & \cdots\cdots & \lambda_m b_{m,1} \\ \vdots & \cdots & \cdots & \cdots \\ \lambda_1 b_{1,i} & \cdots & \cdots & \lambda_m b_{m,i} \\ \vdots & \cdots & \cdots & \cdots \\ \lambda_1 b_{1,m} & \cdots & \cdots & -\lambda_m \end{bmatrix} \tag{6}$$

The independent fission yield Y_i of nuclide *i* at t = 0 in the matrix form is

$$Y_i = \begin{bmatrix} Y_1 \\ Y_2 \\ \vdots \\ \vdots \\ Y_m \end{bmatrix} \tag{7}$$

The number of nuclides *N*(*t*) after shutting down the reactor can be evaluated by solving equation (4) numerically using the fourth order *Runge-Kutta* method, *RKM*, for initial value problem of the ordinary differential equations of the decay system, $N_i'(t) = f(t, N_i(t))$, [16, 17]. 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 this algorithm is in literatures [18-20].

The decay heat power of curium isotopes in a nuclear fuel can be calculated as;

$$f(t) = \sum_i \overline{E}_i^{\beta,\gamma,\alpha}\ \lambda_i\ N_i(t) \tag{8}$$

where, $f(t)$ is the power function (MeV/fission/s) following time t after a fission burst, $\overline{E}_i^{\beta,\gamma,\alpha} = \left(\overline{E}_{\beta,i} + \overline{E}_{\gamma,i} + \overline{E}_{\alpha,i}\right)$, $\overline{E}_{\beta,i}$, $\overline{E}_{\gamma,i}$ 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.1.

The code consists of three subroutines, one of them is to read the initial conditions and extract the branching ratios and decay constant matrix from equations 6 and 7 for all isotopes in the data base, the second one is to solve the ordinary differential equation systems to evaluate the number of nuclides $N(t)$, whereas, the last subroutine is to calculate the decay heat using equation 8.

III. Results And Discussion

The present approach has been applied for decay heat calculation for ^{244}Cm and ^{245}Cm. Figures (1 and 2) show the calculated β-, γ- and total decay heat data using the present approach (*PST*) for fast induced neutron fission, respectively. The results show that at 10 and 1000 seconds after the shutting down the reactor, the highest total decay heat for those isotopes are (0.992 and 1.083) and (0.815 and 0.814) MeV/fission/s, respectively. The contribution of each nuclide to the total FP decay heat following time after fission process for nuclides which contribution larger than 1% is calculated at different cooling time 10 s and 1000 s, for ^{244}Cm and ^{245}Cm and presented in Figs. 3 and 4. The tabulated results in tables 1, show that the highest contributors for ^{244}Cm – fast and ^{245}Cm – thermal are (^{100}Nb and ^{108}Tc) and (^{108}Tc and ^{104}Tc), respectively. Whereas, the least contributors for the two isotopes are (^{140}Xe and ^{102}Mo) and (^{145}Ba and ^{102}Mo), respectively.

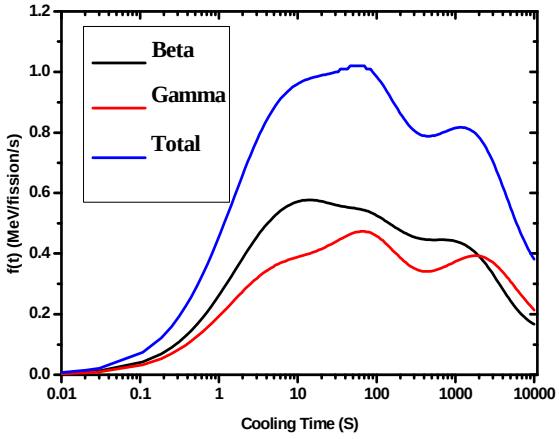

Fig. 1 Beta-, gamma-, and total decay heat for ^{244}Cm- fast induced neutron fission

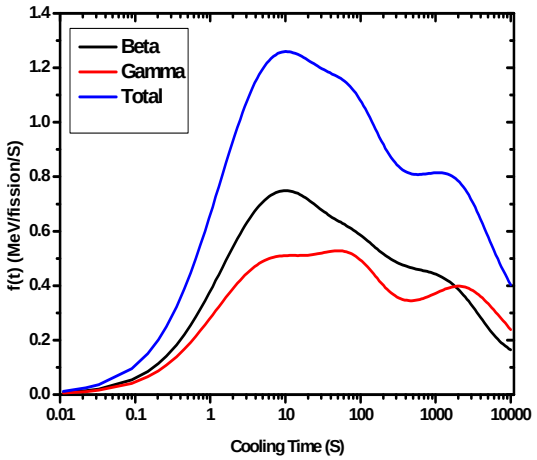

Fig. 2 Beta-, gamma-, and total decay heat for ^{245}Cm- fast induced neutron fission

IV. Conclusions

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.

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TABLE I
DECAY HEAT CONTRIBUTIONS FOR ^{244}CM AND ^{245}CM AT 10 AND 1000 S

^{244}Cm				^{245}Cm			
10 S		1000 S		10 S		1000 S	
Nuclide	Contribution %	Nuclide	Contribution %	Nuclide	Contribution %	Nuclide	Contribution %
100 Nb	5.17	104 Tc	10.04	108 Tc	4.63	104 Tc	10.25
101 Nb	5.08	102 Tc	5.93	100 Nb	4.56	102 Tc	5.49
96 M1 Y	3.06	101 Mo	5.42	110 Rh	4.16	105 Tc	5.40
106 Tc	2.93	139 Cs	5.37	101 Nb	4.15	139 Cs	5.19
102 M1 Nb	2.90	138 Cs	5.33	111 Rh	2.61	101 Mo	5.06
108 Tc	2.84	105 Tc	5.24	143 Ba	2.33	138 Cs	4.95
143 Ba	2.54	141 Ba	4.35	106 Tc	2.28	141 Ba	3.93
92 Rb	2.43	138 Xe	3.54	107 Tc	2.15	138 Xe	3.57
99 Nb	2.28	95 Y	3.49	112 Rh	2.12	143 La	3.25
110 Rh	2.22	143 La	3.46	102 M1 Nb	2.06	108 Rh	3.17
141 Cs	2.14	94 Y	3.12	141 Cs	2.06	142 Ba	2.87
97 Y	2.08	142 Ba	2.97	102 Nb	2.05	134 I	2.85
107 Tc	2.07	93 Sr	2.77	96 Y	1.98	95 Y	2.78
96 Y	2.00	108 Rh	2.25	106 Mo	1.86	133 Te	2.26
100 Zr	1.96	147 Pr	2.19	100 Zr	1.77	93 Sr	2.18
93 Rb	1.86	146 Pr	2.03	93 Rb	1.72	147 Pr	1.88
98 Nb	1.80	137 Xe	2.02	99 Nb	1.68	137 Xe	1.86
145 La	1.76	134 I	1.99	97 Y	1.63	94 Y	1.86
144 La	1.71	89 Rb	1.79	92 Rb	1.60	107 Rh	1.80
102 Nb	1.67	101 Tc	1.79	104 Nb	1.55	146 Pr	1.70
106 Mo	1.62	107 Rh	1.66	146 La	1.51	131 Sb	1.69
140 Cs	1.52	142 La	1.64	105 Mo	1.48	101 Tc	1.68
105 Mo	1.50	107 Ru	1.35	150 Pr	1.40	107 Ru	1.46
103 Mo	1.49	131 Sb	1.26	140 Xe	1.38	142 La	1.45
95 Sr	1.44	133 Te	1.24	135 Te	1.36	89 Rb	1.43
150 Pr	1.34	102 Mo	1.07	144 Ba	1.33	111 Pd	1.34
146 La	1.31	152 Pm	0.99	138 I	1.32	108 Ru	1.23
104 Nb	1.29	151 Nd	0.96	145 La	1.31	152 Pm	1.15
111 Rh	1.28			144 La	1.31	134 Te	1.11
0 M1 Nb	1.23			96 M1 Y	1.28	130 Sb	1.00
6 M1 La	1.14			139 Xe	1.28	102 Mo	0.99
137 I	1.13			95 Sr	1.22		
139 Xe	1.10			103 Mo	1.16		
144 Ba	1.01			149 Ce	1.15		
140 Xe	1.00			98 Nb	1.13		
138 I	0.97			140 Cs	1.10		
136 M1 I	0.95			110 Ru	1.10		
				145 Ba	1.07		
				107 Mo	1.03		

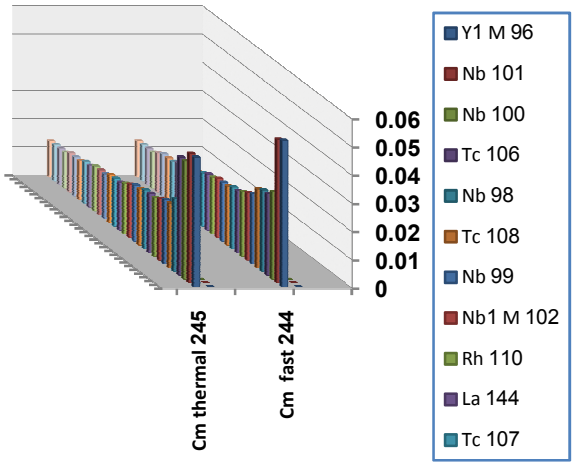

Fig. 3 Main contributions to the total decay heat from Cm- isotopes, calculated at a cooling times 10 s by the present approach

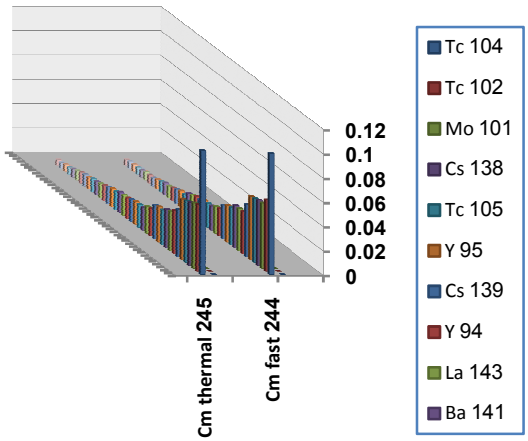

Fig. 4 Main contributions to the total decay heat Cm- isotopes, calculated at a cooling times 1000 s by the present approach

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