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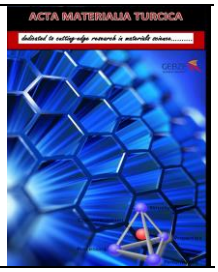

Numerical Estimation of Nuclear Decay Heat from Induced Neutron Fission of ^{235}U and ^{239}Pu

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Abstract:

In this paper, Joint Evaluated Fission and Fusion (JEFF) Nuclear Data Library has been used to calculate the nuclear decay heat after a fission burst of 235U and 239Pu for shutdown time up to 105 sec. This estimation is based on the numerical solution of the linear differential equations that describe buildups and decays of the fission products. The code was written in MATLAB, which is a fast and easy-access platform. The verification of the current code is carried out by comparing the numerical results with the measured reported ones. The discrepancies between the evaluated results and the measured ones show the reliability of the current calculation.

Keywords: Fission yield, Nuclear Decay Heat, Nuclear Data Library, JEFF3.1.

DOI:

1. INTRODUCTION

The heavy nucleus of mass number greater than 130 may undergo a fission reaction when it captures a neutron. However, thermal neutrons (low-energy neutrons) can cause fission only in nuclei with (Z-even and N-odd). Those nuclei such as 235U and 239Pu are called “fissile nuclei”. Whereas for nuclei with (Z-even and N- even), the fission may take place, only if the incident neutrons have energy more than 1 MeV, and the neutrons are called fast neutrons. In those cases, the nuclei are called fissionable.

In the fission process, we can define the "fission yield" as the probability of measuring the production of any nuclide given in production per unit fission. The independent fission yield "Y" is defined as the number of atoms of a specific nuclide produced directly per 100 fission reactions. There are over 1000 fission products that have been identified for all fissionable nuclides such as 235U and 239Pu [1]. Their decays must be tracked for reactors shut-down safety analysis, and for the storage and handling of spent fuel, where both natures of radiations and energy released must be known.

The energy released in the fission process can be divided into two components. The first one is the energy released in the initial fission process, which includes the kinetic energy of the fission fragments, the kinetic energy of the prompt neutrons, and the energy carried off by prompt gamma rays. Meanwhile, the second component so-called “Decay heat”, which is the heat released from fission-product decay, and it is as high as 7.5% of the total energy released in the fission. It decreases slowly and has a large contribution after stopping the neutron chain

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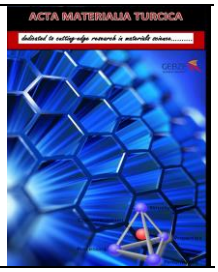

reaction. The insufficient heat removal may cause fuel melting [2, 3].

An accurate calculation of the decay heat as a function of time is important, for many applications, such as safety assessments of nuclear power plants to prevent overheating of reactor fuel following power plant shutdown, handling the fuel during transportation and storage, and management of the radioactive waste[4].

HEATKAU V1, CCC-805, the RSICC code package [5] written in MATLAB environment, where data can be managed easily and visualizable. This code introduced a new approach [6, 7, 8] to evaluate the decay heat power after a fission burst of a fissile nuclide for short cooling time based on the numerical solution of Bateman differential equations [9]. It uses the numerical technique, Runge-Kutta method [10] to solve Bateman equations, to get the number of nuclides "N" after cooling time "t". Then the decay heat power H(t) is calculated for gamma, beta. All the required input data including fission yields and decay data were extracted from both ENDF/ B-VII.0 and ENDF/ B-VII.1 databases.

In the present work, we use HEATKAU code with some modifications. The input data were extracted from Joint Evaluated Fission and Fusion JEFF 3.1 database. Instead of numerical technique Runge-Kutta that was used in HEATKAU, the numerical Backward Differentiation Formulae is used to solve Bateman equations.

The ODE MATLAB solvers that use Runge-Kutta method (of orders two and three or four and five) as ODE23 or ODE45 are a generally useful method only for problems that are not excessively "stiff", but if the problem represents high stiffness, it is advised to use the stiff ODE solver that uses Backward Differentiation Formulae method, ODE15s instead [11, 12].

2. METHODOLOGY

A schematic diagram for the computational model is shown in Fig.1. As shown in the figure, the first step is to calculate the independent fission product distribution with time, N(t). Once N(t) is calculated, the second step in the model is straight forward. The decay heat, H(t) will be calculated by summing of the products of the nuclear activities by the mean alpha, beta and gamma energies released per disintegration of every nuclide in the chain.

The differential equation system that describes the abundances of nuclides in a decay chain as a function of time was reported by Bateman[9] and depends on the initial abundances and decay rates as shown in (Eq.1):

$$\frac{d}{dt}N_i(t) = -\lambda_i N_i(t) + \sum_j b_{j\to i}\lambda_j N_j(t) + Y_i \qquad (2.1)$$

where Ni(t) is the number of nuclides i at a cooling time t , λi is the decay constant of the i-th nuclide, b_(j→i) is the branching ratio to nuclide i per decay of nuclide j, and Y_i is the independent fission yield of nuclide i.

The matrix form of Bateman's equations in (Eq.1) with total number of nuclides "m" can be written in the form of (Eq.2):

$$N' = [B - EYE]*\lambda*N + Y \qquad (2.2)$$

where:

$$\begin{bmatrix} \frac{d}{dt}N_1(t) \\ \frac{d}{dt}N_2(t) \\ \vdots \\ \frac{d}{dt}N_{m-1}(t) \\ \frac{d}{dt}N_m(t) \end{bmatrix} \qquad (2.3)$$

B is the branching ratios matrix that indicates all branching ratios of all nuclides in all chains depending on the modes of decay that mentioned in Table.1:

$$B = \begin{pmatrix} 0 & b_{2\to 1} & \cdots & b_{m\to 1} \\ b_{1\to 2} & 0 & \cdots & b_{m\to 2} \\ \vdots & \vdots & \ddots & \vdots \\ b_{1\to m} & b_{2\to m} & \cdots & 0 \end{pmatrix} \qquad (2.4)$$

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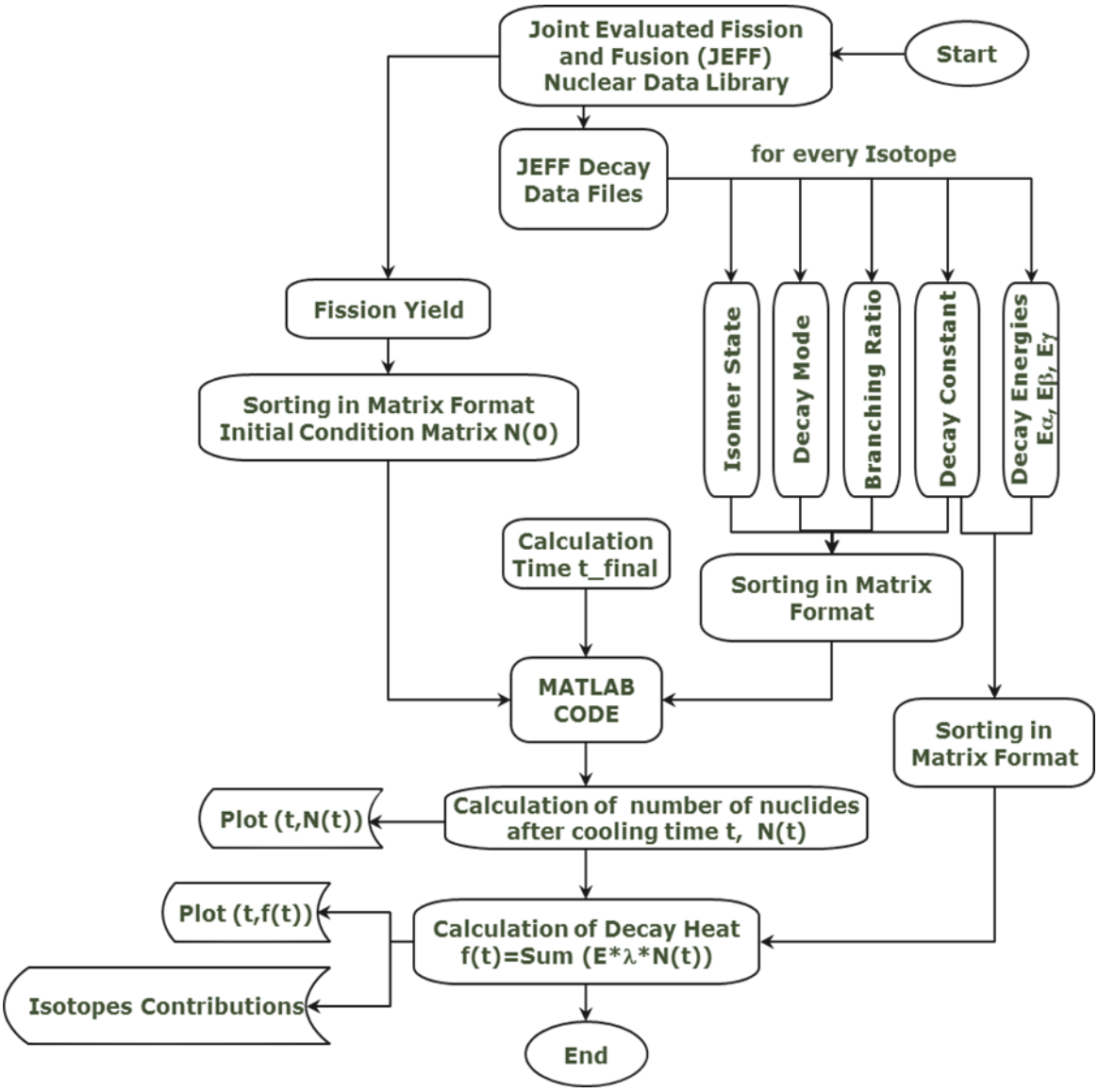

Figure 1. A schematic diagram of the computational model

EYE is the identity matrix as the following:

$$\begin{pmatrix} 1 & 0 & \cdots & 0 \\ 0 & 1 & \ldots & 0 \\ \vdots & \vdots & \ddots & \vdots \\ 0 & 0 & \cdots & 1 \end{pmatrix} \quad (2.5)$$

The matrix form of the decay constant λ can be represented as:

$$\lambda = \begin{bmatrix} \lambda_1 \\ \lambda_2 \\ \vdots \\ \lambda_{m-1} \\ \lambda_m \end{bmatrix} \quad (2.6)$$

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The independent fission product distribution with time, N(t), which is the required solution is:

$$N = \begin{bmatrix} N_1(t) \\ N_2(t) \\ \vdots \\ N_{m-1}(t) \\ N_m(t) \end{bmatrix} \quad (2.7)$$

Y is the matrix of initial independent fission yield:

$$Y = \begin{bmatrix} N_1(0) \\ N_2(0) \\ \vdots \\ N_{m-1}(0) \\ N_m(0) \end{bmatrix} \quad (2.8)$$

The matrices equations (4), (6) and (8) are the input matrices of the code, they were extracted from JEFF3.1 Nuclear Data Library.

The present code was written in MatlabR2019b. ODE15s solver capability was used to solve the above equations numerically by Backward Differentiation Formulae method. ODE15s was chosen to solve initial value problems of high stiffness in a finite time interval [ti - tf] as has been recommended in [13].

The total decay heat can be calculated by (Eq. 9):

$$H(t) = H_\alpha(t) + H_\beta(t) + H_\gamma(t) \quad (2.9)$$

where $H_\alpha(t)$, $H_\beta(t)$,and $H_\gamma(t)$ are alpha, beta, and gamma decay heat respectively and given by equations 10, 11, and 12 respectively.

$$H_\alpha(t) = \sum_{i=1}^{m} \lambda_i N_i(t) E_i^\alpha \quad (2.10)$$

$$H_\beta(t) = \sum_{i=1}^{m} \lambda_i N_i(t) E_i^\beta \quad (2.11)$$

$$H_\gamma(t) = \sum_{i=1}^{m} \lambda_i N_i(t) E_i^\gamma \quad (2.12)$$

Table 1. Possible decay modes used in the present model

Symole	Decay Mode	Daughter nucleus (A,Z)
γ	γ-ray	(A,Z)
β	Beta decay	(A, Z+1)
e.c.,or β +	Electron capture and/or positron emission	(A,Z-1)
IT	Isomeric transition when the state being is an isomeric state	(A,Z)
α	Alpha decay	(A-4,Z-2)
n	Neutron emission	(A-1,Z)
p	Proton emission	(A-1,Z-1)
β −,n	Beta decay followed by neutron emission	(A-1,Z+1)
β −,α	Beta decay followed by alpha emission	(A-4,Z-1)
β +,α	Positron decay followed by alpha emission	(A-4,Z-3)
β +,p	Positron decay followed by proton emission	(A-1,Z-2)
β −, β −	Multiple Beta decay	(A,Z+2)
n,n	Multiple neutron emission	(A-2,Z)
p,p	Multiple proton emission	(A-2,Z-2)
SF	Spontaneous fission	into two nearly equal fragments (found only in very heavy nuclei)

3. RESULTS and DISCUSSION

Alpha, beta, gamma, and total decay heats from thermal-induced neutron fissions of two fissile nuclides ^{235}U and ^{239}Pu have been calculated by the present approach. Calculated data have been compared to the measured ones that were measured in Oak Ridge National Laboratory: ORNL [14,15,16, 17].

Figures 2 and 3 show that the decay heat due to alpha decay of fission products is very small compared to the total decay heat for both 235U and 239Pu thermal fissions. Moreover, figures 4 and 6 show gamma, beta, and total decay heat from thermal-induced neutron fissions of 235U and 239Pu, respectively compared to the measured ones. Figures 5 and 7 show the discrepancies between the calculated gamma, beta, and total decay heat from the measured ones for thermal-induced neutron fissions of 235U and 239Pu respectively, calculated by (Eq.13). The range of discrepancies is reported in table 4.

$$\Delta\% = \left(\frac{PST - Measured}{PST}\right) x100 \quad (3.1)$$

In addition, tables 2 and 3, show the contribution of the most 30 nuclides that affect the total decay heat of the fission products following the time after fission burst, at cooling times 10, 100, and 1000 seconds, for ^{235}U and ^{239}Pu respectively.

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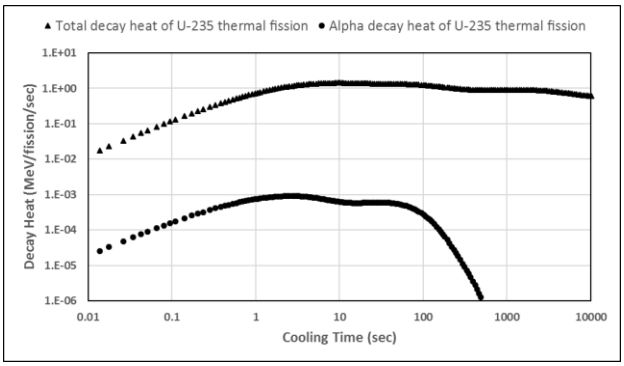

Figure 2. Alpha and total decay heats as a function of time of ^{235}U thermal-induced fission.

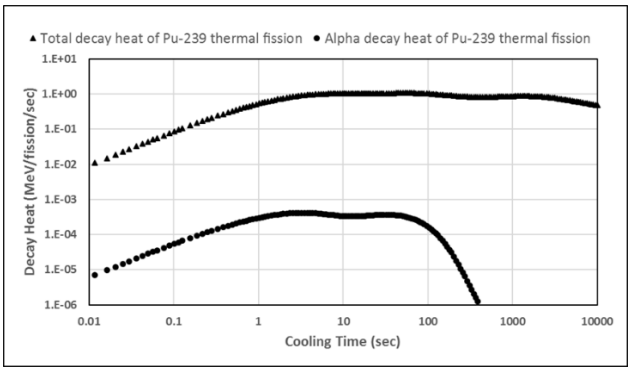

Figure 3. Alpha and total decay heats as a function of time of ^{239}Pu thermal-induced fission.

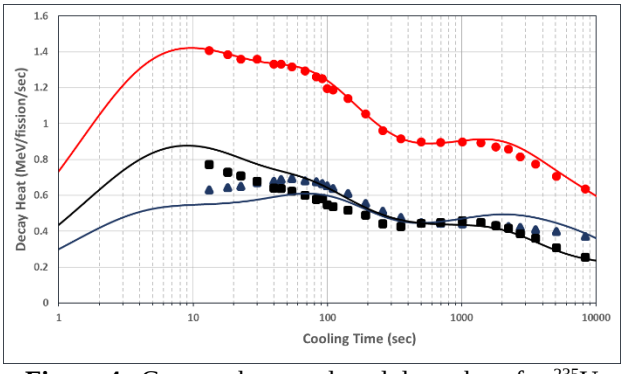

Figure 4. Gamma, beta, and total decay heat for ^{235}U thermal-induced fission. Solid line stands for the present work, whereas, circles, squares, triangles represent the measured total, beta, and gamma decay heat, respectively.

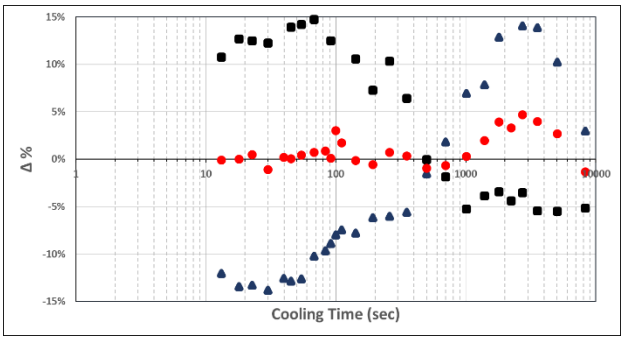

Figure 5. The discrepancies Δ% between the calculated gamma, beta, and total decay heat for ^{235}U thermal-induced fission and the measured ones. Circles, squares, triangles represent measured total, beta, and gamma decay heat, respectively.

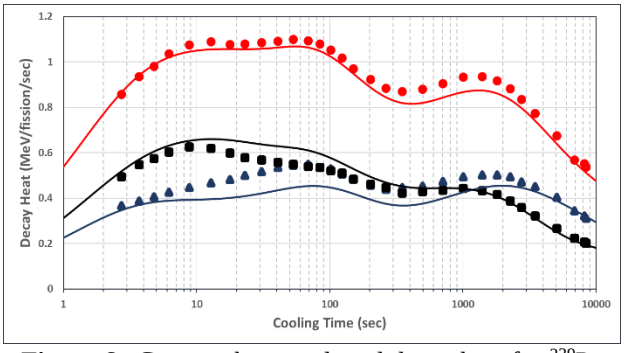

Figure 6. Gamma, beta, and total decay heat for ^{239}Pu thermal-induced fission. Solid line stands for the present work, whereas, circles, squares, triangles represent the measured total, beta, and gamma decay heat, respectively.

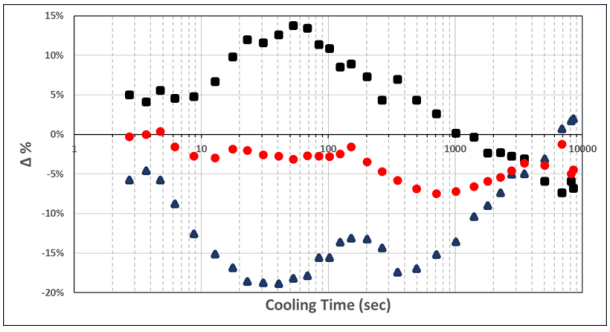

Figure 7. The discrepancies Δ% between the calculated gamma, beta, and total decay heat for ^{239}Pu thermal-induced fission and the measured. Circles, squares, triangles represent measured total, beta, and gamma decay heat, respectively.

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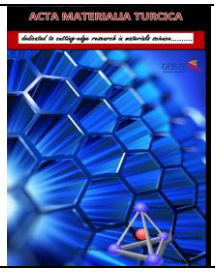

Table 2. Nuclides contribution to the total decay heat for 235U thermal-induced fission calculated at different cooling times by the present work

10 sec		100 sec		1000 sec	
Isotope	**% contribution**	**Isotope**	**% contribution**	**Isotope**	**% contribution**
^{92}Rb	6.26	^{140}Cs	7.53	^{93}Sr***	6.96
^{100M1}Nb	5.68	^{91}Rb	7.05	^{89}Rb	6.87
^{96}Y	4.65	^{144}La	6.70	^{95}Y	6.80
^{93}Rb*	4.41	^{139}Xe	4.28	^{94}Y	6.52
^{143}Ba	3.41	^{136}I	4.24	^{139}Cs	5.25
^{91}Kr	3.31	^{94}Sr	4.11	^{138}Xe	4.52
^{101}Nb	3.27	^{90}Rb	4.09	^{138}Cs	4.43
^{95}Sr	2.69	^{86}Br	3.44	^{101}Mo	4.07
^{102}Nb	2.36	^{87}Br*	2.99	^{141}Ba	4.04
^{140}Xe	2.28	^{136M1}I	2.94	^{143}La	3.60
^{144}Ba	2.26	^{89}Kr	2.93	^{102}Tc	3.57
^{100}Zr	2.23	^{90}Kr***	2.92	^{142}Ba	3.41
^{96M1}Y	2.06	^{98}Nb	2.83	^{133}Te	2.84
^{88}Br*	1.97	^{95}Sr	2.28	^{104}Tc	2.57
^{141}Cs	1.97	^{137}Xe	2.07	^{131}Sb***	2.06
^{135}Te	1.94	^{93}Sr***	2.07	^{137}Xe	1.91
^{145}Ba	1.86	^{141}Cs	1.88	^{134}Te	1.75
^{97}Y	1.85	^{103}Mo	1.81	^{147}Pr	1.74
^{99}Zr* **	1.85	^{133}Sb***	1.60	^{134}I	1.74
^{139}Xe	1.78	^{137}I*	1.45	^{142}La	1.65
^{90}Kr* **	1.74	^{132}Sb	1.44	^{146}Pr	1.63
^{146}La	1.60	^{95}Y	1.29	^{89}Kr	1.44
^{137}I*	1.59	^{103}Tc	1.27	^{101}Tc	1.36
^{138}I*	1.56	^{145}La	1.25	^{90M1}Rb**	1.34
^{98}Nb	1.55	^{145}Ce	1.21	^{90}Rb	1.33
^{99}Nb	1.49	^{99M1}Nb**	1.04	^{133M1}Te**	1.33
^{89}Br*	1.33	^{90M1}Rb**	1.02	^{130}Sb	1.04
^{136}Te	1.25	^{98}Zr	1.02	^{84}Br	0.91
^{94}Rb*	1.22%	^{139}Cs	0.98%	^{130M1}Sb	0.87%
^{91}Rb	1.21%	^{147}Ce	0.97%	^{146}Ce	0.83%

All isotopes in the table are pure beta emitters except those which undergo decay by β$^-$ and β$^-$ followed by neutron (*), those which decay by β$^-$ and isomer transition (**), and those which decay by β$^-$ to daughter ground state and β$^-$ to daughter isomer state

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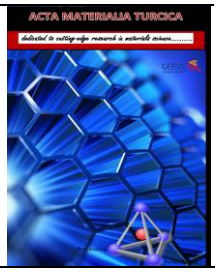

Table 3. Nuclides contribution to the total decay heat for 239Pu thermal-induced fission calculated at different cooling times by the present work

10 sec		100 sec		1000 sec	
Isotope	**% contribution**	**Isotope**	**% contribution**	**Isotope**	**% contribution**
^{100M1}**Nb**	6.87	^{140}Cs	6.34	^{104}Tc	8.65
^{101}**Nb**	4.72	^{106}Tc	5.00	^{95}Y	5.26
^{96}**Y**	3.72	^{144}La	4.97	^{102}Tc	5.25
^{92}**Rb**	3.23	^{103}Mo	4.78	^{101}Mo	5.06
^{96M1}**Y**	3.13	^{136M1}I	3.88	^{139}Cs	4.89
^{143}**Ba**	2.98	^{103}Tc	3.51	^{105}Tc	4.54
^{93}**Rb***	2.77	^{139}Xe	3.38	^{94}Y	4.54
^{100}**Zr**	2.67	^{98}Nb	3.28	^{138}Cs	4.36
^{95}**Sr**	2.25	^{91}Rb	3.18	^{93}Sr***	4.24
^{98}**Nb**	2.15	^{94}Sr	3.03	^{138}Xe	3.70
^{102}**Nb**	2.15	^{136}I	2.83	^{141}Ba	3.67
^{102M1}**Nb**	2.12	^{104}Mo	2.82	^{142}Ba	2.86
^{99}**Nb**	2.09	^{105}Mo	2.69	^{143}La	2.80
^{99}**Zr*** **	2.03	^{137}Xe	2.60	^{89}Rb	2.53
^{106}**Tc**	1.98	^{95}Sr	1.61	^{133}Te	2.25
^{141}**Cs**	1.87	^{141}Cs	1.50	^{131}Sb***	2.25
^{139}**Xe**	1.63	^{93}Sr***	1.49	^{134}I	2.22
^{108}**Tc**	1.59	^{90}Rb	1.46	^{137}Xe	2.00
^{106}**Mo**	1.55	^{86}Br	1.39	^{101}Tc	1.69
^{144}**Ba**	1.55	^{105}Tc	1.37	^{147}Pr	1.65
^{105}**Mo**	1.54	^{132}Sb	1.34	^{133M1}Te**	1.52
^{135}**Te**	1.47	^{99M1}Nb**	1.23	^{142}La	1.48
^{136M1}**I**	1.46	^{95}Y	1.20	^{130}Sb	1.45
^{137}**I***	1.45	^{98}Zr	1.18	^{146}Pr	1.42
^{107}**Tc**	1.42	^{109}Rh	1.17	^{130M1}Sb	1.38
^{97}**Y**	1.41	^{89}Kr	1.15	^{134}Te	1.18
^{140}**Xe**	1.33	^{139}Cs	1.15	^{108}Rh	1.04
^{144}**La**	1.24	^{145}Ce	1.14	^{107}Rh	0.96
^{100M1}**Nb**	1.23	^{102}Tc	1.14	^{102}Mo	0.95
^{104}**Nb***	1.20%	^{137}I*	1.12%	^{107}Ru	0.78%

All isotopes in the table are pure beta emitters except those which undergo decay by β^- and β^- followed by neutron (*), those which decay by β^- and isomer transition (**), and those which decay by β^- to daughter ground state and β^- to daughter isomer state

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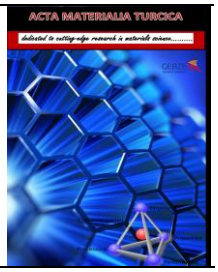

Table 4. Range of Discrepancies between calculated and measured values of gamma, beta, and total decay heat of thermal-induced fission of ^{235}U and ^{239}Pu

	^{235}U thermal-induced fission			**^{239}Pu thermal-induced fission**		
	Gamma	Beta	Total	Gamma	Beta	Total
Range of Discrepancy	-13.8% to 16%	-5.5% to 16.3%	-1.3% to 4.7%	-18.9% to 2%	-7.4% to 13.7%	-7.5% to 0.4%

4. CONCLUSIONS

The numerical evaluation of the number of nuclides after a certain cooling time, using MatlabR2019b, with ODE Ode15s solver capability, led to a good estimation for the decay heat of fissile nuclides. The absolute discrepancies from the measurements were less than 4.7 % and 7.5 % for the total decay heat for induced thermal neutron fission of 235U and 239Pu, respectively. This can be attributed to some missing β-strength of the fission products. The tabulated nuclides in Tables 2 and 3 have the greatest contribution to the total decay heat. This can be of great important for deciding which nuclides need precise measurements. The calculation of alpha decay heat using the present model, shows that it contributes less than 0.0001% to the total decay heat for both fissile nuclides, which is consistent with the literatures. Moreover, it was noted that the major source in the decay heat after fission burst is due to pure beta emitter radioisotopes produced from the decay of the fission fragments. In addition, HEATKAU code has been updated with the data extracted from JEFF 3.1 library. HEATKAU V.II ENDF/B-VIII.0 and JEFF-3.3 libraries will be submitted to RSICC in the near soon.

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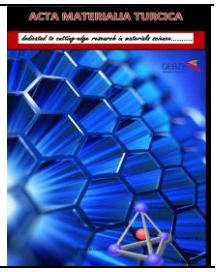

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