Fission Product Decay Heat Calculations for Neutron Fission of $^{232}$Th

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Abstract. Precise information on the decay heat from fission products following times after a fission reaction is necessary for safety designs and operations of nuclear-power reactors, fuel storage, transport flasks, and for spent fuel management and processing. In this study, the timing distributions of fission products’ concentrations and their integrated decay heat as function of time following a fast neutron fission reaction of $^{232}$Th were exactly calculated by the numerical method with using the DHP code.

1. Introduction
In a fission reaction, fission products (FP) are initially formed with known concentrations as independent fission yields, but this data still changes following times after the fission events. This is mainly because of the natural decay of radioactive fission products. In nuclear science and technology, the concentrations of fission products as functions of decay time are the key data required in aggregate decay heat calculations for designs and operations of nuclear-power reactors, fuel storage, transport flasks, and for spent fuel management and processing [1, 2]. Gamma and beta decay energies released from natural decay of the fission products contribute approximately 7% to 12% of the total energy generated through the fission process; this component of power is called “Decay Heat”. After a reactor is shutdown, this source of radioactive decay energy still remains and cumulatively increases that necessary to apply a heat removal system for maintaining the safety level of temperature inside the reactor core.

In nuclear reactor technology, thorium is considering as a potential fuel material that could possibly supplement or even replace natural uranium [3]. $^{232}$Th is the naturally-occurring isotope with abundance of nearly 100%, and can be used as breeding-fuel material by capturing neutrons in a thermal and epithermal neutron flux to form $^{233}$Th isotope and subsequently decay to produce uranium-233 ($^{233}$U), which is an excellent fissile material for nuclear energy production [4]. For fast reactors, in which thorium-232 is used as the breeding-fuel component that driven by a fast neutron spectrum, $^{232}$Th is also fissionable with incident neutrons with energy above one MeV; and the decay energies of its fission products would significantly contribute to the decay heat power in the spent fuel. Accordingly, the updated knowledge of timing distributions or behaviour of concentrations and decay heat energies by fission products from fission reaction of $^{232}$Th is essential necessary in research and application of fast reactor technology.

In this work, the DHP program [5], a numerical calculation code, was used for calculation of fission products decay heat data for fast neutron fission of $^{232}$Th. The method used in this calculation is analysis procedure on the general solutions of the Bateman’s Equations [6] for every full complex decay chain, in which the real-time build-up and decay of FP nuclides are determined. Based on the
decay and fission yield data from JENDL 4.0 [7], the concentration of each FP nuclide is determined as functions of cooling time after a fission event. The difficulties of the complex systems of the decay-chains are solved by an additional computational algorithm implemented in the DHP code.

2. Analysis of Decay and Build-up Numbers

The number of $i^{th}$ nuclide at cooling time $t$ after a fission burst can be calculated from the following equation:

$$N_i(t) = N_i(0) \exp(-\lambda_i t) + \sum_{j=0}^{M} N_{j\to i}(t)$$

in which, $N_i(0)$ is equal to the independent fission yield of the $i^{th}$ nuclide; the component of $N_{j\to i}(t)$ is the build-up number of nuclide $i^{th}$ at cooling time $t$, that was formed in the system of decay chains originated from the nuclide $j^{th}$. This term of the build-up number can be obtained by the general solutions of the Bateman’s equation for every particular linear decay chain. In this work, we developed a numerical algorithm to calculate the term of $N_{j\to i}(t)$ in equation (1) directly by using the decay and fission yield data from JENDL4.0 [7] and/or the evaluated nuclear structure data file ENSDF [8]. The calculation procedure is generalized as the following steps [9]:

- For every nuclide $j^{th}$, the decay net that started from the nuclide $j^{th}$ is separated to form equivalent sub-branches of linear decay chains.

- For each linear decay chain, if the nuclide $i^{th}$ is a daughter nuclide in the decay chain, apply the general solution of Bateman’s equation to calculate the build-up number of nuclide $i^{th}$ due to the decay of nuclide $j^{th}$ through the current linear decay chain, as of the cooling time $t$.

- The total number $N_i(t)$ can be obtained by summing all the build-up numbers of nuclide $i^{th}$ from all the linear decay chains in which the build-up numbers are calculated by apply the general solution of Bateman’s equation [6] for every linear decay branch in a differential period of time $\Delta t$. Let consider to a linear decay branch of $n$ nuclides:

$$N_1 \to N_2 \to N_3 \rightarrow ... N_i \rightarrow ... N_n$$

The build-up number of nuclide $N_i$ due to the decay of nuclide $N_{i-1}$ is calculated by the differential equation:

$$\frac{dN_i(t)}{dt} = \lambda_{i-1}N_{i-1}(t) - \lambda_i N_i(t)$$

(2)

In case of $\{N_i(t) \neq 0 \text{ and all of the remain nuclides } N_{1,2,...j-1,j+1,...M}(t) = 0 \text{ at } t = 0\}$, the solution of build-up number for $N_i(t)$ contributed from nuclide $N_j$ at $t > 0$ is express as:

$$N_{j\to i}(t) = \sum_{m=j}^{i} C_m e^{-\lambda_{m} t}$$

(3)

$$C_m = \frac{\prod_{k=j}^{i} \lambda_k}{\prod_{k=j}^{M} (\lambda_k - \lambda_m)} N_j(0)$$

(4)
In general case of \( \{N_j(t) \neq 0 : j = 1, 2, 3, ... n \text{ at } t = 0 \} \), the total build-up number of time dependent \( N_j(t) \), at \( t > 0 \), contributed from all of other nuclides in a linear decay branch can be estimated as the following general expression.

\[
\sum_j N_{j\rightarrow\ell}(t) = \sum_i \prod_{l=1}^{i-1} N_i(0) \sum_{\nu=1}^i \frac{e^{-\lambda_j}}{\prod_{\ell \neq \nu} (\lambda_{\ell} - \lambda_{\nu})} (5)
\]

A computational procedure for exact calculation of the time-dependent distribution of fission product concentration, \( N_j(t) \), has been developed and integrated in the DHP code. In this calculation, all of decay chains and decay modes including \( \beta^- \) decay to ground state, first and second isomer states, double \( \beta^- \) decay, electron capture decay to ground and isomer states, alpha decay, delay \( \beta^- \) decay, and internal transitions in the fission product system are taken into consider in this calculation. The block diagram of the DHP computational algorithm and window interface are shown in Figure 2 and Figure 3, respectively. The results of calculation for timing concentration functions of several fission product nuclides for times after a fission reaction of \(^{232}\text{Th}\) are shown in Figure 1, in which decay characteristics for every fission product and the independent neutron fission yields were extracted from JENDL4.0 [7].

![Figure 1. Calculated Results of concentration functions for a number selected fission products after a fast neutron fission reaction of \(^{232}\text{Th}\)](image)

3. Beta Decay Average Energies Calculations

The average energy values of beta and gamma-rays released from beta decay of individual fission product are the most important quantities for nuclear decay heat summation prediction. The expressions for average beta and gamma-ray energies based on the Gross theory of beta decay were applied in these calculations [10, 11].
\[
\overline{E}_\beta = T_{1/2} \int_{-\infty}^{0} S_\beta(E) \int_{1}^{m c^2} p E (-E_g + 1 - E)^2 F(E) dEdE_g
\]

(6)

\[
\overline{E}_\gamma = T_{1/2} \int_{-\infty}^{0} S_\beta(E) m c^2 (Q + E_g) \int_{1}^{m c^2} p E (-E_g + 1 - E)^2 F(E) dEdE_g
\]

(7)

where: \( F(E) \) stands for the Fermi function. \( E_g \) is related to the excitation energy \( E_i \) as \( E_g = -(E_i - 1) \); \( m, p \) and \( c \) denotes for electron rest mass, electron momentum and light velocity, respectively. The beta strength function \( S_\beta(E) \) can be determined from the beta feeding function \( I_\beta(E) \) as the following formula:

\[
S_\beta(E) = \frac{I_\beta(E)}{f(Z, Q-E)} \sqrt{T_{1/2}}
\]

(8)

\( f(Z, Q-E) \) is the integrated Fermi function. \( Q, E \) and \( T_{1/2} \) are the beta decay energy, excitation energy in the daughter nuclide and the half-life, respectively. The function \( I_\beta(E) \) can be systematically normalized based on the beta branching intensities adopted from the ENSDF data file, or experimentally derived from measured distributions of beta decay intensity. The calculated results of average beta and gamma energies from beta decay for several selected fission products are shown in Table 1.

**Figure 2.** The block diagram of the computational procedure.
Table 1. The calculated average beta and gamma energies for selected nuclide.

| Nuclide | Q (MeV) | T1/2 (s) | (a) $E_\beta$ (MeV) | (b) $E_\gamma$ (MeV) |
|---------|---------|----------|----------------------|----------------------|
| Rb-89   | 4.496   | 909.0    | 0.9970               | 2.2981               |
| Rb-90   | 6.587   | 158.0    | 2.0512               | 1.9996               |
| Rb-90m  | 6.696   | 265.0    | 1.4047               | 3.2281               |
| Rb-91   | 5.891   | 58.4     | 1.6930               | 2.6675               |
| Rb-93   | 7.462   | 58.4     | 2.5226               | 2.225                |
| Sr-85   | 4.137   | 445.0    | 0.8197               | 2.2709               |
| Sr-85m  | 5.085   | 75.2     | 0.8388               | 1.4503               |
| Sr-90   | 6.087   | 23.9     | 2.2151               | 1.1534               |
| Y-94    | 4.917   | 1120     | 1.4465               | 1.1285               |
| Y-95    | 4.453   | 618.0    | 1.4465               | 1.1285               |
| Cs-138  | 5.374   | 2010.0   | 1.2428               | 2.3646               |
| Cs-138m | 5.547   | 916.0    | 0.2700               | 0.4122               |
| Cs-139  | 4.213   | 556.0    | 1.6567               | 0.3406               |
| Cs-140  | 6.22    | 63.7     | 1.9708               | 1.7946               |
| Cs-141  | 5.251   | 24.9     | 2.0171               | 0.8087               |
| Ba-141  | 3.213   | 1100     | 0.9224               | 0.8679               |
| Ba-142  | 2.211   | 636      | 0.3740               | 1.0852               |
| Ba-143  | 4.246   | 14.3     | 1.2924               | 1.207                |
| Ba-144  | 3.119   | 11.5     | 0.9837               | 0.6239               |
| Ba-145  | 4.923   | 4.31     | 1.4714               | 0.3368               |
| La-142  | 4.503   | 5470.0   | 0.8682               | 2.2952               |
| La-143  | 3.426   | 848.0    | 2.9339               | 0.3468               |
| La-144  | 5.541   | 40.8     | 1.3639               | 2.3128               |
| La-145  | 4.108   | 24.8     | 1.5014               | 0.6509               |
| Ce-145  | 2.535   | 181.0    | 0.6366               | 0.8047               |
| Ce-146  | 1.026   | 811.0    | 0.2543               | 0.3647               |
| Ce-147  | 3.29    | 56.4     | 1.4929               | 0.2469               |
| Ce-148  | 2.06    | 56.0     | 0.5635               | 0.3416               |
| Pr-146  | 4.169   | 1450.0   | 1.3294               | 0.9969               |

Figure 3. The window interface of the DHP program.
The average energies shown in Table 1 are calculated in two cases: (a) calculated based on the input data of branching intensities extracted from the ENSDF data files; (b) calculated from the experimental beta decay intensities measured by Greenwood et al. [12] in which a total absorption gamma-rays spectrometer (TAGS) was used; TAGS is an efficient experimental method for average energies determinations, that overcome the problem of pandemonium effects [13].

4. Results of Calculations
The summation model for decay heat calculations is as the following function:

\[
f(t) = \sum_{i=1}^{M} \frac{E_i}{\lambda_i} N_i(t)
\]

where: \(M\) denotes the maximum number of FP nuclides; \(E_i = E_{i\beta} + E_{i\gamma}\) stands for the mean energy per decay of the \(i\)th nuclide, \(\lambda_i\) the decay constant, \(N_i(t)\) the corresponding concentration function for cooling time \(f(t)\) is the burst function of decay heat \((\text{MeV/Fission/s})\). The physical quantity equal to \(t \times f(t)\) is called decay heat power function \((\text{MeV/Fission})\).

In this work, the JENDL FP Decay Data File 2011 [14] and fission yield data file from JENDL 4.0 [7] have been applied for calculations of fission product concentrations as functions of cooling time after fast neutron fission reactions of \(^{232}\)Th, and these results were adopted in decay heat calculations. The results of calculations for \(t \times f(t)\), as functions of times after a fission event, are shown in Table 2 and Figure 4 in comparison with measured values by Akizama 1982 [15].

| Time after fission burst (s) | Decay Heat \(t \times f(t)\) (MeV/Fission) | Time after fission burst (s) | Decay Heat \(t \times f(t)\) (MeV/Fission) |
|-----------------------------|------------------------------------------|-----------------------------|------------------------------------------|
|                            | Beta | Gamma | Total |                            | Beta | Gamma | Total |
| 1.10E-01                    | 0.1497 | 0.1191 | 0.2688 | 3.00E+02 | 0.4812 | 0.5834 | 1.0646 |
| 1.60E-01                    | 0.2065 | 0.1641 | 0.3706 | 4.00E+02 | 0.4419 | 0.5208 | 0.9627 |
| 2.10E-01                    | 0.2579 | 0.2048 | 0.4627 | 5.00E+02 | 0.4225 | 0.4908 | 0.9132 |
| 2.60E-01                    | 0.3047 | 0.2418 | 0.5464 | 6.00E+02 | 0.4121 | 0.4760 | 0.8880 |
| 3.10E-01                    | 0.3475 | 0.2755 | 0.6231 | 7.00E+02 | 0.4061 | 0.4684 | 0.8745 |
| 4.10E-01                    | 0.4234 | 0.3352 | 0.7586 | 8.00E+02 | 0.4027 | 0.4646 | 0.8673 |
5. Conclusions
In the present work, the computer code DHP has been improved for calculation procedure and updated for new available nuclear databases. Calculations have been carried out for the decay and growth distributions of concentrations for fission products from fast neutron fission of $^{232}$Th. The calculated data of fission product concentrations were then introduced into summation calculations of total and partial decay heat following times after fission burst. The decay and fission yield data used in this work is extracted from JENDL FP Decay Data File and fission yield data file 2011 [14]. As shown in Table 2 and Figures 4, within the decay time period from $1\times10^2$ s to $5\times10^3$ s, the present calculated

| Time (s) | Decay Heat (W/kg) |
|---------|-------------------|
| 1.00E+00 | 0.9978 0.9379 1.0656 2.00E+03 0.3987 0.4677 0.8638 |
| 2.00E+00 | 1.0972 0.8680 1.9452 8.00E+03 0.2912 0.3730 0.6857 |
| 3.00E+00 | 1.0938 0.8969 1.9907 9.00E+03 0.2857 0.3598 0.6455 |
| 4.00E+00 | 1.0967 0.9157 2.0124 1.00E+04 0.2814 0.3473 0.6287 |
| 5.00E+00 | 1.0722 0.8680 1.9452 8.00E+03 0.2912 0.3730 0.6857 |
| 6.00E+00 | 1.0922 0.9281 2.0203 1.50E+04 0.2678 0.2929 0.5607 |
| 7.00E+00 | 1.0838 0.9362 2.0200 2.00E+04 0.2565 0.2516 0.5081 |
| 8.00E+00 | 1.0734 0.9415 2.0148 2.50E+04 0.2446 0.2217 0.4662 |
| 9.00E+00 | 1.0620 0.9447 2.0067 3.00E+04 0.2321 0.1999 0.4320 |
| 1.00E+01 | 1.0610 0.9471 1.9532 4.00E+04 0.2064 0.1707 0.3771 |
| 1.50E+01 | 1.0904 0.9454 1.9058 5.00E+04 0.1815 0.1516 0.3331 |
| 2.00E+01 | 1.0795 0.9473 1.8487 6.00E+04 0.1753 0.1380 0.2972 |
| 3.00E+01 | 1.0529 0.9525 1.8550 7.00E+04 0.1406 0.1278 0.2684 |
| 4.00E+01 | 1.0726 0.9651 1.8377 8.00E+04 0.1255 0.1199 0.2454 |
| 5.00E+01 | 1.0814 0.9721 1.8236 9.00E+04 0.1135 0.1136 0.2270 |
| 6.00E+01 | 1.0838 0.9706 1.8013 1.00E+05 0.1039 0.1083 0.2122 |
| 7.00E+01 | 1.0805 0.9618 1.7702 1.50E+05 0.0767 0.0908 0.1675 |
| 8.00E+01 | 1.0750 0.9477 1.7327 2.00E+05 0.0641 0.0821 0.1461 |
| 9.00E+01 | 1.0712 0.9302 1.6914 2.50E+05 0.0573 0.0785 0.1359 |
| 1.00E+02 | 1.0685 0.9105 1.6482 3.00E+05 0.0539 0.0780 0.1320 |
| 1.50E+02 | 1.0633 0.8039 1.4402 4.00E+05 0.0520 0.0812 0.1332 |
| 2.00E+02 | 1.0543 0.7092 1.2735 5.00E+05 0.0528 0.0861 0.1388 |
| 2.50E+02 | 0.5149 0.6362 1.1511 6.00E+05 0.0544 0.0907 0.1451 |

Figure 4. Calculated fission product decay heat after fast neutron fission of $^{232}$Th
results are agreements with the experimental values measured by Akizama [15]. It is also estimated that the updated DHP code can be used for calculations of fission product inventory concentrations and decay heat data exactly.

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**References**

[1] Nichols A L 2002 “Nuclear Data Requirements for Decay Heat Calculations” Lectures given at the Workshop on Nuclear Reaction Data and Nuclear Reactors: Physics, Design and Safety, Trieste, 2002

[2] Oyamatsu K 1999 “Easy-To-Use Application Programs to Calculate Aggregate Fission Product Properties on Personal Computers” JEARI-Conf 99-002

[3] Kazimi M S, Czerwinski K R, Driscoll M J, Hejzlar P and Meyer J E 1999 “On the Use of Thorium in Light Water Reactors” MIT-NFC-TR-016

[4] OECD 2015 “Introduction of Thorium in the Nuclear Fuel Cycle”, NEA No. 7224

[5] Son P N and Katakura J 2007 “An Application Program for Fission Product Decay Heat Calculations” JAEA-Data/Code 2007-018

[6] Tobias A 1980 Prog. Nucl. Energy 5 1

[7] Shibata K O, et al 2011 J. Nucl. Sci. Technol. 48 1

[8] Bhat M R 1992 “Evaluated Nuclear Structure Data File (ENSDF)”, Nuclear Data for Science and Technology, Edited by Qaim S M, Springer-Verlag, Berlin, Germany, Page 817

[9] Son P N 2014 Int. J. of Nucl. Ener. Sci. and Enginee. 4 75

[10] Katakura J, Yoshida T, Oyamatsu K, and Tachibana T 2001 “JENDL FP Decay Data File 2000” JAERI-1343 (2001)

[11] Yoshida T and Katakura J 1986, Nucl. Sci Eng. 93 193

[12] Greenwood R C, Helmer R G, Putnam M H, and Watts K D 1997 Nucl. Instr. Meth. A390 95

[13] Hardy J C, Carraz L C, Jonson B and Hansen P G 1977 Phys. Letts. 71B(2) 307

[14] Katakura J 2011 “JENDL FP Decay Data File 2011 and Fission Yields Data File 2011” JAEA-Data/Code 2011-025

[15] Akiyama M and An S 1982 “Measurement of fission-product decay heat for fast reactors” Proc. Int. Conf. on Nuclear Data for Science and Technology, Antwerp Belgium (1982): pp 237-244