Analysis of Fission Fragments Contributors on Total Decay Heat of Thermal Neutron-Induced Fission of U-235

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Abstract
Calculation of the decay heat from the decay/buildup of radionuclides generated after nuclear fission is one of the highest priorities in the nuclear industry. These calculations become more important if they are made together with the analysis of the most important isotopes affecting the decay heat. They are useful in designing the necessary nuclear safety for spent fuels, and their importance cannot be overlooked in the designs of transporting fuel storage containers as well as in the management of the radioactive waste generated. In this paper, by using MATLAB, the decay heat after the thermal fission of a U-235 nucleus was numerically calculated by solving linear differential equations for all the buildups/decays of the fission products. Also, the most contribution of radioactive isotopes to the decay heat was analyzed by using Microsoft Excel. The most influential isotopes were deduced in two ways; either by calculating the most influential isotopes at specific times, or by determining the largest influences in a cumulative manner. All required nuclear data such as decay constants their branching ratios, independent fission yield, and average α-, β-, and γ-energies released per disintegration of any nuclide, have been extracted from the latest version of the Evaluated Nuclear Data Files (ENDF) database ENDF/B-VIII.0. The two different methods used showed a difference in the contributing isotopes, which is logical for the difference in the method of calculation. The first method is suitable for instantaneous data while the second method is more suitable when there is a need to know the cumulative calculations. In sum, we can say that both methods complement each other, and neither of them can be dispensed with in the accurate calculations related to transportation and storage of spent fuel.

Keywords
Fission Products, Decay/Buildup, Fission Yield, Decay Heat
1. Introduction

In nuclear power plants, energy is obtained because of the nuclear fissions of some fissionable nuclei, which are usually actinides by thermal or fast neutrons [1]. Actinides are very important in studying the spent nuclear fuel to some extent. In those cases, interest is focused on uranium, plutonium, neptunium, americium, and curium. About 7% of the energy produced from the fission process is in the form of kinetic energies of the fission products, which is so-called “Decay Heat”. It is very important to accurately predict the total heat of dissolution for several purposes, including those related to the design purposes of nuclear plants and ensuring their safety as well as the transportation and storage of spent fuel [2] [3].

Several studies have been concerned with the analysis and study of the radioactive isotopes that most affect the nuclear safety of spent fuel as a result of the decay/buildup of fission products [4]-[12].

The method used to calculate the decay heat in this research is the microscopic summation method [13], in which the decay heat is calculated separately from each of the radioactive isotopes within the fission products. Then the effects of all radioactive isotopes are summed in order to get the decay heat as a result of each type of radiation (Alpha, Beta, and Gamma), which in turn gives us the total decay heat if added.

It is not possible to calculate the decay heat of all radioisotopes without starting by calculating the nuclide concentrations of all isotopes \( N(t) \), where \( t \) represents the elapsed time after the fission event. Softwares with high computational capabilities such as MATLAB or MATHCAD for solving differential equations enable us to use numerical methods without any simplification or approximation, as all decay/buildup series have been determined, and all possible decay modes have been taken into consideration [14]. Nuclear data were specifically extracted from the decay data library and fission product data file of the latest version of the ENDF database ENDF/B-VIII.0.

Decay heat and isotopic concentrations as a function of cooling time are of great importance for the safe handling of spent fuel and also for maximizing the quality of storage and cooling conditions. For safety or design purposes, many researchers are developing the predictive capabilities of fission product isotopic concentrations by designing simulation models capable of identifying and analyzing radioactive isotopes. Therefore, in this paper, two different approaches were used to describe the highest contributed isotopes from fission products influencing the decay heat after the fission event. The first method determines the most effective isotopes emitted from different fission products at specific times regardless of the decay/buildup of these isotopes. Whereas, the second method is more comprehensive that it takes into account the cumulative effect of determining the isotopes that have the highest effect of fission products on the decay heat. The first method gives us the required isotopes at specific times in time, while the second method takes care of the entire time period. The most 20 con-
tributors’ nuclides for the most important fissile actinide are presented. This fissile actinide is uranium-235.

2. Methodology

The processes of calculating the decay heat resulting from the decay of fission products carried out by the present code are shown in the flowchart in Figure 1. The first step is to extract the necessary nuclear data from the ENDF/B-VIII.0 including the fission yield for the specific actinide under study. These data are represented in terms of ordinary differential equations as Initial Value Problem. Also, all the decays of each isotope are extracted to form the decay chains, and thus branching ratios are extracted for each decay in the chain. Half-life and the average energy released in each decay (with branches, if any) are also extracted. These data are extracted for all the fission products (usually more than one thousand) and then all the differential equations are solved for all the isotopes present in addition to those that were not present but resulted from the decay of the fission products. Simply, we can write the basic equation for calculating the

![Figure 1. Flowchart processes of the code.](image-url)
decay heat due to the $i^{th}$ isotope for decomposition products after the fission event $f(t)$ as follows:

$$f(t) = \lambda_i N_i(t) E_i^\alpha + \lambda_i N_i(t) E_i^\beta + \lambda_i N_i(t) E_i^\gamma$$

(1)

where, $E_i^\alpha$, $E_i^\beta$ and $E_i^\gamma$ are the average energies released per disintegration of isotope $i$ of alpha, beta, and gamma, respectively, $\lambda_i$ is the decay constant of isotope $i$, and $N_i(t)$ is the number of $i^{th}$ isotope or nuclide present at time $t$ due to any decay or buildup process.

The 4th order numerical method Runge-Kutta (RK4) [15] is used to solve the decay/buildup fission products, so the inventory $N_i(t)$ of any nuclide can be calculated using the following Equations (2) and (3):

$$\frac{dN_i(t)}{dt} = -\lambda_i N_i(t) + \sum_{j=M}^{j=M} b_{j-i} \lambda_j N_j(t)$$

(2)

$$N_i(0) = Y_i$$

(3)

where $M$ is the number of fission products, $j$ represents all nuclides in which one of the decay products is a nuclide $i$, $b$ is the branching ratio, and $Y_i$ is the independent fission yield of nuclide $i$.

Equation (1) can be rewritten as Equations (4) to (6), whereas, the total decay heat is given by Equation (7) as:

$$f(t)_a = \sum_{i=1}^{i=M} \lambda_i N_i(t) E_i^\alpha$$

(4)

$$f(t)_\beta = \sum_{i=1}^{i=M} \lambda_i N_i(t) E_i^\beta$$

(5)

$$f(t)_\gamma = \sum_{i=1}^{i=M} \lambda_i N_i(t) E_i^\gamma$$

(6)

$$f(t)_{\text{total}} = f(t)_a + f(t)_\beta + f(t)_\gamma$$

(7)

where, $f(t)_a$, $f(t)_\beta$ and $f(t)_\gamma$ are the decay heat released per disintegration of all nonzero $M$ isotopes of alpha, beta, and gamma, respectively, $f(t)_{\text{total}}$ is the total decay heat due to all isotopes for the decomposition products after the fission event. $M$ does not necessarily include all isotopes in the fission products since it excludes stable isotopes, and it also includes unstable isotopes that are generated via fission fragments decay/buildup and are not initially present in the fission products.

3. Results and Discussion

The current code was used to solve Equations (2) and (3) to calculate the inventory of the nuclides present since the beginning of thermal fission $N_i(t)$ for U-235 due to thermal fission. The number of fission products present in this case is 923 isotopes.

After numerically solving the simultaneous 923 differential equations for fission products of U-235 thermal fission (in addition to all the nuclides expected to form after fission and not among the fission products at the beginning), the exact time-dependent distribution of the fission product buildup/decay is calcu-
lated up to a cooling time $t = 10^7$ seconds after the thermal fissions with neutrons of energy $0.0235$ eV. Inventories of some important fission products are shown in Figure 2. The results presented in Figure 2 show the decay/buildup curves of isotopes resulting from the fission products (via thermal fission of U-235 nucleus), plotted on a semi-logarithmic scale to show the decay rate of each isotope clearly. The results show that the short-lived radioactive isotopes decay very quickly in a short time, and most of the important radioactive isotopes are born shortly after the fission event, and some are even long-lived such as cesium-137 (half-life = $9.5 \times 10^8$ seconds). The decay/buildup curves of the fission products are not sufficient on their own to know the most effective isotope in the decay heat unless we combine it with the average energy emitted for each decay of each isotope per second.

The next step in the code is to calculate the decay heats (alpha, beta, and gamma) for each isotope using Equations (4)-(6), respectively. And then by use Equation (7), we calculate the total decay energy for each isotope. Then by adding up the decay heats for all the radioactive isotopes, we can calculate the total decay heat.

Using the numerical solutions of Equations (4) to (7), it is possible to analyze the most active isotopes in the decay heat, and this paper will be concerned with the analysis of isotopes that contribute the overall decay heat by taking advantage of the great analytics capabilities of MATLAB and Microsoft Excel. Two methods have been adopted to analyze these influences or contributors. In the first method, the isotopes were first arranged in descending order according to the most influential isotopes in the decay heat at 3 different times after fission event: 100, 10,000, and 1,000,000 seconds. Then the most 20 contributors on the

![Figure 2](#). Inventory of some important fission products of thermal fission of U-235.
Table 1. Most 20 Contributors on overall decay heat for fission of U-235 due to thermal neutron at 10² sec.

| Nuclide | Z  | A  | Isomer State | Cont % | Nuclide | Z  | A  | Isomer State | Cont % |
|---------|----|----|--------------|--------|---------|----|----|--------------|--------|
| La      | 57 | 144| 0            | 8.02   | Rb      | 37 | 92 | 0            | 3.68   |
| Cs      | 55 | 140| 0            | 7.89   | Nb      | 41 | 100| 0            | 2.92   |
| Rb      | 37 | 91 | 0            | 7.85   | Sr      | 38 | 95 | 0            | 2.79   |
| Sr      | 38 | 94 | 0            | 4.39   | La      | 57 | 144| 0            | 2.64   |
| I       | 53 | 136| 0            | 4.05   | Rb      | 37 | 93 | 0            | 2.52   |
| Xe      | 54 | 139| 0            | 3.81   | Rb      | 37 | 91 | 0            | 2.29   |
| kr      | 36 | 89 | 0            | 3.39   | Zr      | 40 | 99 | 0            | 2.13   |
| Br      | 35 | 87 | 0            | 3.38   | Ba      | 56 | 143| 0            | 2.10   |
| Br      | 35 | 86 | 0            | 3.37   | kr      | 36 | 91 | 0            | 2.04   |
| Nb      | 41 | 98 | 0            | 2.93   | Y       | 39 | 96 | 0            | 1.99   |
| kr      | 36 | 90 | 0            | 2.87   | Cs      | 55 | 140| 0            | 1.97   |
| Sr      | 38 | 95 | 0            | 2.69   | Cs      | 55 | 141| 0            | 1.90   |
| Xe      | 54 | 137| 0            | 2.25   | Nb      | 41 | 102| 0            | 1.89   |
| Sr      | 38 | 93 | 0            | 2.14   | Y       | 39 | 97 | 0            | 1.85   |
| Mo      | 42 | 103| 0            | 2.06   | Y       | 39 | 96 | 1            | 1.82   |
| Cs      | 55 | 141| 0            | 2.03   | Xe      | 54 | 139| 0            | 1.79   |
| La      | 57 | 145| 0            | 1.87   | kr      | 36 | 90 | 0            | 1.75   |
| I       | 53 | 136| 1            | 1.75   | Nb      | 41 | 98 | 0            | 1.74   |
| Sb      | 51 | 133| 0            | 1.73   | Nb      | 41 | 101| 0            | 1.69   |
| Sb      | 51 | 132| 0            | 1.65   | Sr      | 38 | 96 | 0            | 1.66   |
| Total Cont % | 70.09% | Total Cont % | 43.17% |

total decay heat were selected. Results are presented in Tables 1-3. The left parts of these tables are dedicated to calculating the effect of the top 20 nuclides on the total decay heat at the specified times (first method), while the right parts are dedicated to the second method, which calculates the cumulative effect of radioactive nuclides on the total decay heat starting from the fission event until reaching the specified times. For example, in Table 1, the top 20 radioactive nuclides affect more than 70% of the total heat of decay at 100 seconds using the first method, while the second method shows that the top 20 radioactive nuclides contribute only about 43% of the total heat of decay. Also, the three most effective nuclides in the first method are La-144, Cs-140, and Rb-91, while the three most effective nuclides in the second method are Rb-92, Nb-100, and Sr-95. In addition to the above, we find that among the top 20 effective radionuclides calculated by the
Table 2. Most 20 Contributors on overall decay heat for fission of U-235 due to thermal neutron at $10^4$ sec.

| Nuclide | Z  | A  | Isomer | State | Cont % | Nuclide | Z  | A  | Isomer | State | Cont % |
|---------|----|----|--------|-------|--------|---------|----|----|--------|-------|--------|
| I       | 53 | 134| 0      |       | 21.51  | Rb      | 37 | 92 | 0      |       | 2.31   |
| La      | 57 | 142| 0      |       | 12.88  | Cs      | 55 | 138| 0      |       | 2.18   |
| Cs      | 55 | 138| 0      |       | 7.80   | La      | 57 | 144| 0      |       | 2.18   |
| Rb      | 37 | 88 | 0      |       | 6.52   | Rb      | 37 | 91 | 0      |       | 2.16   |
| Sr      | 38 | 92 | 0      |       | 5.71   | Cs      | 55 | 140| 0      |       | 2.05   |
| kr      | 36 | 88 | 0      |       | 4.98   | Sr      | 38 | 95 | 0      |       | 1.85   |
| I       | 53 | 135| 0      |       | 4.70   | Nb      | 41 | 100| 0      |       | 1.83   |
| Ba      | 56 | 139| 0      |       | 4.23   | Sr      | 38 | 93 | 0      |       | 1.76   |
| Y       | 39 | 92 | 0      |       | 3.80   | I       | 53 | 134| 0      |       | 1.72   |
| La      | 57 | 141| 0      |       | 3.36   | Y       | 39 | 94 | 0      |       | 1.58   |
| kr      | 36 | 87 | 0      |       | 3.31   | Rb      | 37 | 93 | 0      |       | 1.58   |
| Te      | 52 | 133| 1      |       | 2.66   | Y       | 39 | 95 | 0      |       | 1.58   |
| Te      | 52 | 134| 0      |       | 2.36   | kr      | 36 | 89 | 0      |       | 1.46   |
| Sr      | 38 | 91 | 0      |       | 2.29   | Rb      | 37 | 89 | 0      |       | 1.44   |
| Y       | 39 | 93 | 0      |       | 1.51   | Xe      | 54 | 139| 0      |       | 1.36   |
| Pr      | 59 | 145| 0      |       | 1.15   | Zr      | 40 | 99 | 0      |       | 1.33   |
| Pr      | 59 | 146| 0      |       | 1.11   | Ba      | 56 | 143| 0      |       | 1.33   |
| Te      | 52 | 131| 0      |       | 0.93   | Sr      | 38 | 94 | 0      |       | 1.31   |
| Zr      | 40 | 97 | 0      |       | 0.91   | kr      | 36 | 91 | 0      |       | 1.28   |
| I       | 53 | 133| 0      |       | 0.88   | Cs      | 55 | 141| 0      |       | 1.27   |

Total Cont % | 92.59% | Total Cont % | 33.56%

two methods at 100 sec, 8 are found in the two methods, namely Cs-140, Cs-141, kr-90, La-144, Nb-98, Rb-91, Sr-95 and Xe-139.

In Table 2, the top 20 radioactive nuclides affect more than 90% of the total heat of decay at 104 seconds using the first method, while the second method shows that the top 20 radioactive nuclides contribute less than 34% of the total heat of decay. Also, the three most effective nuclides in the first method are I-134, La-142, and Cs-138, while the three most effective nuclides in the second method are Rb-92, Cs-138, La-144. In addition to the above, we find that among the top 20 effective radionuclides calculated by the two methods at $10^4$ sec, only 2 are found in the two methods, namely Cs-138, and I-134.

Finally, in Table 3, the top 20 radioactive nuclides represent almost all of the total decay heat, with a percentage of 99.55% at time $10^6$ seconds using the first
Table 3. Most 20 Contributors on overall decay heat for fission of U-235 due to thermal neutron at 10^6 sec.

| Nuclide | Z  | A  | Isomer | Cont % | Nuclide | Z  | A  | Isomer | Cont % |
|---------|----|----|--------|--------|---------|----|----|--------|--------|
| La      | 57 | 140| 0      | 43.84  | I      | 53 | 134| 0      | 2.14   |
| I       | 53 | 132| 0      | 16.95  | Rb     | 37 | 92 | 0      | 2.08   |
| Ba      | 56 | 140| 0      | 6.81   | Cs     | 55 | 138| 0      | 2.08   |
| Pr      | 59 | 143| 0      | 4.42   | La     | 57 | 144| 0      | 1.96   |
| Zr      | 40 | 95 | 0      | 3.95   | Rb     | 37 | 91 | 0      | 1.95   |
| I       | 53 | 131| 0      | 3.86   | Cs     | 55 | 140| 0      | 1.84   |
| Xe      | 54 | 133| 0      | 2.82   | Sr     | 38 | 95 | 0      | 1.66   |
| Sr      | 38 | 89 | 0      | 2.43   | Nb     | 41 | 100| 0      | 1.65   |
| Mo      | 42 | 99 | 0      | 2.40   | Sr     | 38 | 93 | 0      | 1.59   |
| Nd      | 60 | 147| 0      | 2.13   | La     | 57 | 142| 0      | 1.54   |
| Te      | 52 | 132| 0      | 2.05   | Y      | 39 | 94 | 0      | 1.43   |
| Ru      | 44 | 103| 0      | 1.84   | Rb     | 37 | 93 | 0      | 1.42   |
| Ce      | 58 | 141| 0      | 1.81   | Y      | 39 | 95 | 0      | 1.42   |
| Pr      | 59 | 144| 0      | 1.19   | kr     | 36 | 89 | 0      | 1.32   |
| Y       | 39 | 91 | 0      | 1.13   | Rb     | 37 | 89 | 0      | 1.30   |
| Nb      | 41 | 95 | 0      | 0.79   | Xe     | 54 | 139| 0      | 1.23   |
| Ce      | 58 | 143| 0      | 0.53   | Zr     | 40 | 99 | 0      | 1.20   |
| Pm      | 61 | 149| 0      | 0.28   | Ba     | 56 | 143| 0      | 1.19   |
| Sb      | 51 | 127| 0      | 0.20   | Sr     | 38 | 94 | 0      | 1.18   |
| Ce      | 58 | 144| 0      | 0.11   | kr     | 36 | 91 | 0      | 1.15   |

Total Cont % | 99.55% | Total Cont % | 31.33%

method, while the second method shows that the top 20 radioactive nuclides contribute less than 32% of the total heat of decay. Also, the three most effective nuclides in the first method are La-140, I-132, and Ba-140, while the three most effective nuclides in the second method are I-134, Rb-92, and Cs-138. Also, we find that there are no common contributors among the top 20 effective radio-nuclides calculated by the two methods at 10^6 sec.

4. Conclusion

Decay heat is one of the important physical quantities in the field of nuclear engineering. Since its calculation depends on the precession in the nuclear data, the use of the latest versions of nuclear data libraries is crucial. In the present work, these calculations were carried out by extracting all the required nuclear data
from ENDF/B-VIII.0 library. This database contains the independent fission yield data for 31 Actinides. These independent fission yields are the initial values of our initial value problem. Definitely updating the nuclear data increases the accuracy of the results of the study that was conducted based on it, and this applies to each of the decay temperatures and the nuclides affecting them. Both methods used in this research to adopt and analyze the most effective nuclides or contribute to decay heat due to thermal fission of the U-235 nucleus have proven their importance, noting that the two methods do not give the same results. The tabulated results show that the first method used to identify the influent cores is entirely different from the second method at all times chosen. In the first method, at the preset times entered into the code, the software analyzes all the components of the total decay heat of all the influencing nuclides at the specified time only without getting preoccupied with the values before or after that specified time. The code then arranges these nuclides in descending order from highest to least effective. The importance of this method is shown in determining instantaneous measurements. As for the second method, it analyzes and studies the effects from the fission event to the time of measurement cumulatively, and this method is useful for aggregate calculations. Finally, we can say that both methods complement each other, and neither of them can be dispensed with in the accurate calculations related to transportation and storage of spent fuel.

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Conflicts of Interest

The author declares no conflicts of interest regarding the publication of this paper.

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