Decay heating power of various types of plutonium

Zhengming Gao$^{1,3}$, Juan Zhao$^2$

$^1$School of Computer Engineering, Jingchu University of Technology, Jingmen, 448000, China
$^2$ School of Electronics and Information Engineering, Jingchu University of Technology, Jingmen, 448000, China

$^3$E-mail: gaozmjg@jcut.edu.cn

Abstract. Analysis of the influence of the decay heating power of plutonium is one of the important tasks of the process in design of nuclear reactors, nuclear waste disposal or mechanical analysis of nuclear explosive devices. The paper simulated the decay heating power of some isotopes by means of cascade decay dynamics, then numerical regression was done and some formulae were obtained describing the heating power of the isotopes respectively. Furthermore, the paper reached to a common formula describing the heating power of all kinds of plutonium, and the heating powers of various types of plutonium used in fields of nuclear energy, nuclear waste disposal or military were obtained. Comparing with other calculation methods, calculating the heating power of various types of plutonium using this common formula raised in the paper could result in higher precision and more efficient, and because of the time-dependence, the common formula could also be used in thermal-related analysis of plutonium considering the storage time.

1. Introduction

Plutonium is the 94th element in the periodic system of elements, it is near the center of actinide family (which includes elements from 89th to 103rd). It is one of the main fissile materials used in nuclear plants and nuclear warheads, so it is strategically important in nuclear energy and military fields. Plutonium has several isotopes, e.g. the isotopes which the atomic mass is 238, 239, 240, 241 and 242. $^{238}\text{Pu}$, $^{239}\text{Pu}$, $^{240}\text{Pu}$, $^{241}\text{Pu}$ and $^{242}\text{Pu}$ are commonly used in fields of nuclear energy and nuclear weapons, they are the main components of the plutonium material used in these areas. Each of the five isotopes is radioactive and emits heat and light. Our former study indicates that the heat produced in storing plutonium is mainly the cascade decay heat [1]. Hereby, calculation of the cascade decay heat is one of the inevitable process in the work when designing nuclear reactors, disposing nuclear waste and analyzing the mechanical characteristics of nuclear warheads.

To the authors’ knowledge, there are two ways to calculate the cascade decay heat when needed. One is to calculate it precisely according to the fissile data library. Percentage of $^{238}\text{Pu}$ varies violently in nuclear plants and processing of nuclear waste, which leads the cascade decay heat to change with it. To the technologists working in such area, nuclear data library is accessible and professional software can directly provide the result, so precise work can be done [2, 3]. But it needs a lot of work to obtain the data and program to other technologists who have no such software at hand. So there is another way to acquire the decay heat when needed, which is the simplified calculation considering the main component (e.g. professor Xiao calculated the decay heat of $^{239}\text{Pu}$ to represent the heat of the weapon-grade plutonium in his temperature simulation procedure of nuclear explosive devices [4]) or just...
adding the decay heat of each isotope with weights of their percentage [5]. The simplified calculation method saves a lot of work and the results are adequately used in estimation. But such results cannot be suitable when the objects change with time.

We conducted this study to obtain a common formula calculating the decay heat of various types of plutonium. With such formula, technologists working in fields of nuclear plants, nuclear waste disposal and design of nuclear weapons can derive the decay heat of the specific plutonium used in their studies with high precision and time-relevance.

2. Decay heating power of plutonium isotopes

$^{238}\text{Pu}$, $^{239}\text{Pu}$, $^{240}\text{Pu}$, $^{241}\text{Pu}$ and $^{242}\text{Pu}$ are the common isotopes used in nuclear energy and military area. $^{238}\text{Pu}$ and $^{241}\text{Pu}$ of them are two isotopes with small half time, their atoms have higher probability to decay, whereas other isotopes of the five ones have large half time and their atoms have lower probability to decay. We split the cascade series of each isotope into one or more single cascade series, then we calculate the atomic number $N_i(t)$ of each isotope when it is one kilogram initially with the cascade decay dynamics [6]. Therefore, the degree of activity of the $i$th element is:

$$A_i = \lambda_i N_i(t)$$  

(1)

Concluding from the definition of the degree of activity, the decay heat under specific conditions is:

$$Q_i = E_iA_i = E_i\lambda_i N_i(t)$$  

(2)

Adding the heat of each single cascade series of the isotopes, we derive the decay heat of the specific isotope when it decays from a mass of one kilogram initially.

2.1. Decay heating power of $^{238}\text{Pu}$

First of all, we inquire the decay parameters of $^{238}\text{Pu}$ and her daughters from a public nuclear data library [7], then, we calculate the decay heating power of each isotope in the decay series with the cascade decay dynamics and equation (2). Summing up all of the heat power of the mother isotope and the daughters, the decay heat of $^{238}\text{Pu}$ cascade decay series when pure $^{238}\text{Pu}$ is one kilogram in mass initially is derived and their contribution ratios are also clear (seen in Fig. 1). Considering the storage of plutonium in real world, 200 years is the upper limit in simulation. In Fig. 1, the longitude coordinate $y_i$ is a function of the contribution ratio of the ($i$th-1) generation daughter as:

$$y_i = \log\left(\frac{q_i}{\sum_{j=1}^{n} q_j}\right)$$  

(3)

where $Q_i$ or $q_i$ is the decay heating power of the $i$th -1 or $j$th -1 generation daughter.

The decay heating power HPP$_{238}(t)$ is mainly decided by the decay heat of the mother in 200 years and the contribution ratio to HPP$_{238}(t)$ of each daughter raises themselves with time (Fig. 1). It is significant of the change of contribution of the first daughter $^{234}\text{U}$. We can also find that the decay heat of other isotopes except $^{234}\text{U}$ and the mother contribute lower than $10^{-4}$ in order of magnitude to the power. So the standard error is less than $1\%_o$ for the decay heating power when considering purely the decay heat of the mother and the first generation daughter.

Polynomial regression is done to the calculated data of $^{238}\text{Pu}$ cascade decay series and the result is:

$$\text{HPP}_{238} = \begin{cases} 0.0084t^2 - 3.8515t + 558.4518 & 0 < t \leq 200 \\
568.1474 & t = 0 \end{cases} \text{ (Watt)}$$  

(4)

where $t$ has a unit in years as follows.

Causing the large error and the common use when $t = 0$, the decay heating power of $^{238}\text{Pu}$ is divided into two segments. Test of significance is done according to the following reliable function [8]:

$$\text{HPP}_{238} = \begin{cases} 0.0084t^2 - 3.8515t + 558.4518 & 0 < t \leq 200 \\
568.1474 & t = 0 \end{cases} \text{ (Watt)}$$  

(4)
Fig. 1. Decay heating power of $^{238}\text{Pu}$ and contribution-ratios of its daughters

$$SSR = \sum_{i=1}^{n} \left( y_{i} - \bar{y} \right)^{2}, \quad SSI = \sum_{i=1}^{n} \left( y_{i} - \bar{y} \right)^{2}, \quad R = \frac{SSR}{SSI} \quad (5)$$

In equation (5), $y_{i}$ is the calculated data of decay heating power, $\bar{y}$ is their mean value and $\hat{y}$ is the regressive power. The result is $R = 0.9993$, which means the degree of belief is 99.93%, so equation (5) is suitable to describe the heating power of $^{238}\text{Pu}$ cascade decay series.

2.2. Decay heating power of $^{239}\text{Pu}$, $^{240}\text{Pu}$, $^{241}\text{Pu}$ and $^{242}\text{Pu}$

The same work can be done to calculate the heat producing power of $^{239}\text{Pu}$, $^{240}\text{Pu}$, $^{241}\text{Pu}$ and $^{242}\text{Pu}$ and similar results can be obtained. All of them and their first generation daughters except that of $^{240}\text{Pu}$ cascade decay series are the main contributors to the decay heating power. Because of the large half-life of $^{240}\text{Pu}$ or its low decay probability, the decay heating power of $^{240}\text{Pu}$ is mainly composed of that of the mother. The standard error is less than 1%, so simplified calculating of the decay heating power when only the mother and the first generation daughter are considered is useful in thermal related studies in analysing nuclear explosive devices and thus a lot of work is saved.

The results of the curve of the decay heating power of $^{239}\text{Pu}$, $^{240}\text{Pu}$, $^{241}\text{Pu}$ and $^{242}\text{Pu}$ are shown in Fig. 2, Fig. 3, Fig. 4 and Fig. 5. Polynomial regressions are done to the calculated power and we derived the following equations:

$$HPP_{239} = -5.532 \times 10^{-5}t + 1.9299 \quad (6)$$

$$HPP_{240} = -7.7695 \times 10^{-4}t + 7.4371 \quad (7)$$

$$HPP_{241} = \begin{cases} 
4.3963 \times 10^{-9}t^5 - 2.7190 \times 10^{-6}t^4 + 6.5051 \times 10^{-4}t^3 \\
-0.0756t^2 + 4.1662 + 15.9299 \\
12.9286 
\end{cases}, \quad 0 < t \leq 180 \quad (8)$$

$$t = 0$$

$$HPP_{242} = 0.1170 \quad (9)$$

The degree of belief derived from equation (5) is 99.99999%, 99.9999%, 99.98% and 99.9999999% which told us that those equations from (6) to (9) are reliable.
2.3. Comparison and analysis
Reference value of the decay heating power of $^{238}$Pu is 560 W/Kg [9] or 567 W/Kg [10]. Contrast with equation (5), it is obvious that they were the decay heating power when $t = 0$, and one of them is smaller probably due to the difference of nuclear data.

Reference values of the decay heating power of $^{239}$Pu, $^{240}$Pu, $^{241}$Pu and $^{242}$Pu are 1.9W/Kg, 6.8W/Kg, 4.2 W/Kg and 0.1 W/Kg [9, 10], which are almost the same as our results except that of $^{241}$Pu. The reference values are the decay heat of the isotopes. The little difference between the reference values and the calculated data indicates that the contribution to the decay heat power of the mother is very high. Comparison between the reference value and the calculate result of $^{241}$Pu shows that decay heat of $^{241}$Pu contributes less to the decay heating power, whose numeral value is 32.5% when $t = 0$.

3. Decay heating power of plutonium
The plutonium used in military and nuclear energy are all mainly composed of the five isotopes [11, 12]. The five equations (4) and (6) to (9) show that if their weighted percentage in the specific plutonium are known, then we can reach to the decay heating power of the plutonium, which is the weighted sum of percentage and the five equations.

3.1. Common formula of the decay heating power of plutonium
If the weighted percentage of $^{238}$Pu, $^{239}$Pu, $^{240}$Pu, $^{241}$Pu and $^{242}$Pu in a specific plutonium is $m_1$, $m_2$, $m_3$, $m_4$ and $m_5$ respectively, then adding equations (4) and (6) to (9) weighted the percentage, we get an equation:
\[
\begin{align*}
    P &= \begin{cases} 
    4.3963 \times 10^{-9} t^5 m_4 - 2.7190 \times 10^{-6} t^4 m_4 \\
    + 6.5051 \times 10^{-4} t^3 m_4 + (0.0084 m_1 - 0.0756 m_4) t^2 \\
    - (3.8515 m_1 + 5.532 \times 10^{-5} m_2 + 7.7695 \times 10^{-4} m_3 - 4.1662 m_4) t \\
    + (558.4518 m_1 + 1.9299 m_2 + 7.4371 m_3 + 15.9299 m_4 + 0.1170 m_5) \\
    \end{cases} & 0 < t \leq 180 \\
    \end{align*}
\]

The decay heating power of plutonium described by equation (10) depends on the time (in year) and weighted percentages of the components.

### 3.2. Decay heating power of specific types of plutonium

Plutonium used in military and nuclear plants can be divided into five types [9]: super-grade plutonium, weapon-grade plutonium, reactor-grade plutonium, mixed oxide (MOX)-grade plutonium and FBR blanket, as shown in Tab. 1.

#### Table 1. Various types of plutonium and their components

| grade        | \(^{238}\text{Pu}\) | Percentage in weight | \(^{239}\text{Pu}\) | \(^{240}\text{Pu}\) | \(^{241}\text{Pu}\) | \(^{242}\text{Pu}\) |
|--------------|-----------------|---------------------|-----------------|-----------------|-----------------|-----------------|
| Super-grade  | -               | 0.98                | 0.02            | -               | -               | -               |
| Weapons-grade| 0.00012         | 0.938               | 0.058           | 0.0035          | 0.00022         | -               |
| Reactor-grade| 0.013           | 0.603               | 0.243           | 0.178           | 0.078           | -               |
| MOX-grade    | 0.019           | 0.404               | 0.321           | 0.178           | 0.078           | -               |
| FBR blanket  | -               | 0.96                | 0.04            | -               | -               | -               |

Summing up equations (4) and (6) to (9) with weights of the percentages tabled in Tab. 1, we can reach to the equations calculating the decay heating power per kilogram of specific types of plutonium:

\[
P_{\text{Supu}} = -6.9753 \times 10^{-5} t^2 + 2.0400 \\
P_{\text{WgPu}} = \begin{cases} 
    1.5387 \times 10^{-11} t^5 - 9.5165 \times 10^{-9} t^4 + 2.2768 \times 10^{-6} t^3 \\
    -2.6359 \times 10^{-4} t^2 + 0.0014 t + 2.3644 \\
    2.3551 \\
    \end{cases} & 0 < t \leq 180 \\
    \end{align*}
\]

\[
P_{\text{RgPu}} = \begin{cases} 
    -5.6883 \times 10^{-12} t^5 + 4.1955 \times 10^{-9} t^4 - 1.2589 \times 10^{-6} t^3 \\
    -1.9822 \times 10^{-4} t^3 - 0.0017 t^2 + 0.7716 t + 12.7102 \\
    12.6673 \\
    \end{cases} & 0 < t \leq 180 \\
    \end{align*}
\]

\[
P_{\text{MOXpu}} = \begin{cases} 
    7.8254 \times 10^{-10} t^5 - 4.8398 \times 10^{-7} t^4 + 1.1579 \times 10^{-4} t^3 \\
    -0.0013 t^2 + 0.6681 t + 16.6222 \\
    16.2722 \\
    \end{cases} & 0 < t \leq 180 \\
    \end{align*}
\]

\[
P_{\text{FBRPu}} = -8.4185 \times 10^{-5} t^2 + 2.1502 \\
\]

### 3.3. Application and advantages

The common formula, equation (10), and the specific equations (11) to (15) are time dependent and accurate to calculate the decay heating power of various types of plutonium used in fields nuclear energy, nuclear waste disposal and design of nuclear weapons. With such formulae, we can reach to precise results of a specific plutonium with only the percentages value in mass of each isotopes, then large error of simplified calculation [4, 5] can be avoided. Causing the formulae are time dependent, they can be used in time relevant analysis, e.g. transient thermal analysis of the nuclear explosive
 devices. The decay heating power of the specific plutonium can also be determined whereas the uncertainty statement of “15 to 30 watts” in the thermal analysis of plutonium storage container [13].

4. Results and discussion

Considering the inconvenience or even impossible to gain relevant software which can compute the decay heating power of plutonium automatically, we have the anxiety to find a way easy to do so with as more accurate as it can. Most of jobs relevantly simply computed the decay heating power at the beginning. In order to gain better and accurate results when the plutonium were stored for some time, the composed decay heating power would be computed considering the mothers and their first level daughters. In such circumstances, the mean errors of the decay heating power might be reduced to 1‰. However, the errors would increase along with time. Consequently, when the plutonium used in plants, especially in military, they would be stored for a long period before the final usage. Under such conditions, the errors might be quite large and would make differences, even result in a false conditions. Therefore, the decay heating power of plutonium along with time is needed literally.

The cascade decay dynamics is of course the inherent logical way to calculate the decay heating power. We here finally, reach to a formula which can calculate the decay heating power of all kinds of plutonium composing the five isotopes studied and specific formulae due to specific types of plutonium used in fields of military, nuclear plants and nuclear waste disposal are also given in the end.

5. Conclusions

In this paper, we derived a formula that can compute the decay heating power of plutonium with accuracy and time-relevant. We can also use it to calculate the decay heating power of various types of plutonium used in laboratory, experiments, plants, or even military. It would be precise and widely useful of the common formula (10) to calculate the decay heating power. The results can be used in specific thermal analysis of various types of plutonium or their containers.

Acknowledgements

The authors would like to thank the supports of the following projects:
1) Hubei Provincial Natural Science Foundation with grant number 2019CFB661;
2) The cultivatable science foundations of Jingchu University of Technology with grant number PY201903;
3) The key research and development project of Jingmen with grant number 2019YFZD009;
4) The research project of Hubei Provincial Department of Education with grant number B2019213;
5) The general Excellent Students Work Funding Project of Hubei Provincial Colleges with grant number 2019XGJPB3013;
6) The second batch of scientific research team of Jingchu University of Technology with grant number TD202001.

References

[1] Office of Non-proliferation Research and Engineering 2001 Technology R&D for Arms Control [R] Livermore: Lawrence Livermore National Laboratory
[2] Bennett C H, Steck J E, Behrman E C 2000 Quantum information and computation [J] Nature 404 (3) 247-255
[3] De-cheng QU, Yong-fu CHANG 2004 A Recursive Algorithm of Multi-Branches Decay Chains [J] Atomic Energy Science and Technology 1 (1) 1-4 (In Chinese)
[4] Gang XIAO, Wen-kai ZHU 2005 Numerical Simulation for Three Dimensional Temperature Fields of Composite Body with Plutonium-239 [J] ACTA Armamentar II 1 (1) 140-142 (In Chinese)
[5] Cheng-an LIU, Jun WU 2007 An Introduction of Verification Technology of Nuclear Arms Control [M] Beijing: National Defense Industry Press 4 14
[6] Yan-sen WANG, Fu-ting SHI 1998 Nuclear Physics [M] Beijing: Atomic Energy Press 12 364-378 (In Chinese )

[7] Y S Cheng, R A Guilmette, Y Zhou, et al. 2004 Characterization of plutonium aerosol collected during an accident [J] Health Phys. 87 (6) 596-605

[8] R. Kouzes, B. Geelhood 2002 Composite Signatures of Nuclear and Non-Nuclear Technologies for Weapons Material and Component Measurement [R] Washington, USA: Pacific Northwest National Laboratory 4

[9] Richard L. Garwin 1994 Explosive Properties of Various Types of Plutonium: NATO Advanced Research Workshop "Managing the Plutonium Surplus: Applications and Options" London [C] Chatham House 1 24

[10] Amory B. Lovins 1980 Nuclear weapons and power-reactor plutonium [J] Nature 1.28 283 (5750) 817-823

[11] Siegfried S. Hecker 2000 Plutonium and Its Alloys -From atoms to microstructure [J] Los Alamos Science 26 290-335

[12] Siegfried S. Hecker 2000 Mechanical Behaviour of Plutonium and Its Alloys [J] Los Alamos Science 26 336-355

[13] Saso Stevkovski 1999 Thermal Analysis of Plutonium Storage Containers [D] Texas: Texas Tech University 8