Study on Characteristic of Temperature Coefficient of Reactivity for Plutonium Core of Pebbled Bed Reactor

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Abstract. As a part of the solution searching for possibility to control the plutonium, a current effort is focused on mechanisms to maximize consumption of plutonium. Plutonium core solution is a unique case in the high temperature reactor which is intended to reduce the accumulation of plutonium. However, the safety performance of the plutonium core which tends to produce a positive temperature coefficient of reactivity should be examined. The pebble bed inherent safety features which are characterized by a negative temperature coefficient of reactivity must be maintained under any circumstances. The purpose of this study is to investigate the characteristic of temperature coefficient of reactivity for plutonium core of pebble bed reactor. A series of calculations with plutonium loading varied from 0.5 g to 1.5 g per fuel pebble were performed by the MCNPX code and ENDF/B-VII library. The calculation results show that the $k_{\text{eff}}$ curve of 0.5 g Pu/pebble declines sharply with the increase in fuel burnup while the greater Pu loading per pebble yields $k_{\text{eff}}$ curve declines slightly. The fuel with high Pu content per pebble may reach long burnup cycle. From the temperature coefficient point of view, it is concluded that the reactor containing 0.5 g-1.25 g Pu/pebble at high burnup has less favorable safety features if it is operated at high temperature. The use of fuel with Pu content of 1.5 g/pebble at high burnup should be considered carefully from core safety aspect because it could affect transient behavior into a fatal accident situation.

Keywords: temperature coefficient of reactivity, plutonium core, pebble bed reactor, MCNPX, ENDF/B-VII

1. Introduction

The superiority in quantity of resources and compatibility with environment makes nuclear energy becomes the most prospective energy among the various energy resources in overcoming the problem of increase in the world energy demand. Thirteen countries and institutions in the world that pursue the field of nuclear energy technology have been collaborating in the Generation IV international forum (GIF) with the aim of increasing the role of nuclear energy systems in helping meet the world energy needs in the future. Some Generation IV reactors with characteristic of using fuel more
efficiently, reducing waste production, being economically competitive, and fulfilling stringent standards of safety and proliferation resistance will be demonstrated in the next decade, with commercial deployment beginning in 2030 [1,2].

The pebble bed reactor, a type of the HTGR which uses graphite as neutron moderator and helium as coolant, was selected as the most promising one of the six Generation IV nuclear reactors. This is because the reactor is designed to have a high operating temperature to produce high energy conversion efficiency supporting the application of industrial process heat such as hydrogen production, and others [3,4]. The pebble bed reactor is also designed to have the flexibility that offers the possibility of using fuel with a series of cycling to reduce the accumulation of plutonium resulted from a nuclear power plant (NPP) and military activities. The growing accumulation of plutonium clearly raises public concern about the radiotoxicity dangers and also opportunities to use plutonium not for peaceful purposes [5].

As a part of the solution searching that possible to control the plutonium, a current effort is focused on mechanisms to maximize consumption of plutonium. Plutonium core solution is a unique case in the high temperature reactor which is intended to reduce the accumulation of plutonium [6]. However, the performance of plutonium core which tends to produce a positive temperature coefficient of reactivity should be examined. The pebble bed inherent safety features which are characterized by a negative temperature coefficient of reactivity must be maintained under any circumstances.

The purpose of this study is to investigate the characteristic of temperature coefficient of reactivity for plutonium core of pebble bed reactor. A series of calculations were performed by the MCNPX [7] code and ENDF/B-VII library [8]. The calculation results of temperature coefficient with plutonium loading varied from 0.5 g to 1.5 g per fuel pebble were discussed to complete the analysis.

2. Pebble Bed Reactor
Pebble bed reactor has a cylindrical core with a diameter of 3 m and height of 9.43 m connected to a cone at the bottom of the reactor core with height of 0.61 m. A power density of 3 MW/m$^3$ generates reactor thermal power of 200 MW. The reactor operates at helium inlet and outlet temperatures of 550 ºC and 950 ºC, respectively, with pressure of 4 MPa and the mass flow rate of 120 kg/s. The reactor parameters and core specifications of pebble bed reactor are given in table 1.

| Table 1. Reactor parameter and core specification of pebble bed reactor [9]. |
|----------------------------------|------------------|
| **Reactor parameter**            |                  |
| Thermal power (MW)               | 200              |
| Core diameter/height (m/m)       | 3/9.43           |
| Helium inlet/outlet temperature (°C) | 550/950         |
| Helium pressure (MPa)            | 5.2              |
| Helium flowrate (kg/s)           | 120              |
| Helium density (g/cm$^3$)        | 1.78×10$^4$      |

| **Core speification**            |                  |
|----------------------------------|------------------|
| Core specific power (MW/m$^3$)   | 3                |
| Number of fuel pebble per m$^3$  | 5,394            |
| Number of fuel pebble in core    | 359,548          |
| Packing fraction                 | 0.61             |
| Natural boron impurity in graphite structure (ppm) | 2               |

The reactor core is loaded by 359,548 fuel pebbles with a packing fraction of 0.61 while the cone is filled with 17,263 moderator pebbles with the same packing fraction. Packing fraction is defined as the ratio of total pebble to core volume. Helium coolant flows from top to bottom of the core through the space between pebbles arranged randomly. Fuel pebble and moderator pebble have the same diameter of 6 cm, but different material content. Moderator pebble is made of pure graphite
with density of 1.84 g/cm$^3$. Fuel pebble consists of two regions: the first region, which is called the fueled-zone, 5 cm in diameter containing thousands TRISO particles dispersed in graphite matrix and the second region, which is called a graphite shell, surrounds the fueled-zone with 0.5 cm thick. Graphite matrix and graphite shell act as neutron moderator while moderator pebble as an extra moderator in the reactor core.

The TRISO particle is made of a plutonium oxide (PuO$_2$) kernel with a density of 10.4 g/cm$^3$ and it is wrapped by four coating layers: porous carbon buffer layer, inner pyrolytic carbon (iPyC) layer, a layer of silicon carbide (SiC) and outer pyrolytic carbon (oPyC). The fuel kernel has plutonium isotopic vector as shown in Table 2. The four coating layers protect and preserve effectively the metallic and gas fission product from releasing to the environment for the temperatures up to 1,600 ºC. The overall diameter of TRISO particle is 660 cm. The pebble fuel specifications are summarized in Table 3.

### Table 2. Plutonium isotopic vector [10].

| Isotope | Pu vector (%) |
|---------|---------------|
| $^{238}$Pu | 2.59         |
| $^{239}$Pu | 53.85        |
| $^{240}$Pu | 23.66        |
| $^{241}$Pu | 13.13        |
| $^{242}$Pu | 6.77         |

### Table 3. Fuel pebble specification of pebble bed reactor [10].

| Fuel pebble                  |       |
|------------------------------|-------|
| Pebble diameter (cm)         | 6     |
| Fueled-zone diameter (cm)    | 5     |
| Graphite shell thickness (cm)| 0.5   |
| Graphite shell density (g/cm$^3$) | 1.75   |
| Graphite matrix density (g/cm$^3$) | 1.75   |
| Natural boron impurity in graphite shell (ppm) | 1.0 |
| Natural boron impurity in graphite matrix (ppm) | 1.0 |

| TRISO coated particle         |       |
|------------------------------|-------|
| Fuel kernel                  | PuO$_2$ |       |
| Diameter (µm)               | 240     | 40    |
| Density (g/cm$^3$)           | 10.4    | 1.90  |
| Natural boron impurity in kernel (ppm) | 1.0 |
| SiC layer                    | Thickness (µm) | 35 |
| Density (g/cm$^3$)           | 3.18    |

| Buffer layer                 |       |
|------------------------------|-------|
| Thickness (µm)               | 95     |
| Density (g/cm$^3$)           | 1.05   |

During reactor operation, the fuel pebble can pass through the core to be recirculated several times in Multi-pass loading scheme, or only once in the once-through-then-out (OTTO) loading scheme. The mechanism of fuel loading in OTTO scheme is simple, so it is not necessary to have the equipment for fuel burnup measurement, fuel recirculation and extraction equipment to cancel the fuel to be reused. Multipass scheme requires all those mechanisms. The fuel which is not reused will be sent to the spent fuel storage tank for further processing.
3. Calculation Model

One of main factors in the pebble bed reactor modeling is feature of double heterogeneity characterized by randomness of TRISO coated particles distributed in the fuel pebble (first heterogeneity) and randomness of fuel pebbles distributed in the reactor core (second heterogeneity). MCNPX as a general purpose Monte Carlo radiation transport code has capability to model this double heterogeneity in detail and accurately. The modeling of pebble bed reactor consists of model of TRISO particles in fuel pebble and model of fuel pebbles in core region.

3.1. TRISO Particles Model

The randomness of TRISO particles in pebble was modeled by a simple cubic (SC) lattice. Each PuO\textsubscript{2} kernel with its four coating layers was placed in the center cubic lattice. This cubic lattice is defined as UNIVERSE. Fuel pebble was then modeled by applying LATTICE and FILL options with repetitive structures provided by MCNPX.

In this study, the content of plutonium per pebble was varied from 0.5 g to 1.5 g. Plutonium content (\(m_{Pu}\)) in fuel pebble is defined by the equation,

\[
m_{Pu} = \frac{4\pi}{3} r_{kernel}^3 \rho_{Pu} \times N_{TRISO}
\]

where,

- \(\rho_{Pu}\) is plutonium density,
- \(N_{TRISO}\) is number of TRISO particles per pebble, and
- \(r_{kernel}\) is radius of the fuel kernel.

The calculation of the plutonium density (\(\rho_{Pu}\)) follows the equation,

\[
\rho_{Pu} = \frac{A_{Pu}}{A_{Pu} + 2A_O} \times \rho_{PuO2}
\]

where,

- \(\rho_{PuO2}\) is PuO\textsubscript{2} density,
- \(A_{Pu}\) is plutonium atomic weight, and
- \(A_O\) is oxygen atomic weight.

The calculation of the plutonium atomic weight (\(A_{Pu}\)) is undertaken by the equation,

\[
A_{Pu} = \left[ \frac{\alpha_{Pu238}}{A_{Pu238}} + \frac{\alpha_{Pu239}}{A_{Pu239}} + \frac{\alpha_{Pu240}}{A_{Pu240}} + \frac{\alpha_{Pu241}}{A_{Pu241}} + \frac{\alpha_{Pu242}}{A_{Pu242}} \right]^{-1}
\]

where,

- \(A_{Pu238}, A_{Pu239}, A_{Pu240}, A_{Pu241}\) and \(A_{Pu242}\) are atomic weight of \(^{238}\)Pu, \(^{239}\)Pu, \(^{240}\)Pu, \(^{241}\)Pu and \(^{242}\)Pu, respectively, and
- \(\alpha_{Pu238}, \alpha_{Pu239}, \alpha_{Pu240}, \alpha_{Pu241}\) and \(\alpha_{Pu242}\) are isotopic vector of \(^{238}\)Pu, \(^{239}\)Pu, \(^{240}\)Pu, \(^{241}\)Pu and \(^{242}\)Pu, respectively.

Plutonium content per pebble can be determined by changing the kernel radius where the number of TRISO particles is kept constant or by changing the number of TRISO particle in the fuel pebble where the kernel radius is kept constant. In this calculation, the kernel radius with standard size is preserved at fixed value. To get a different plutonium loading with specification from 0.5 to 1.50 g, the number of TRISO particles in the fuel pebble was changed from 7,526 to 22,577. The changed
TRISO particle number causes the size of SC lattice pitch \( p \) should be adjusted following the equation,

\[
p = r_{\text{zone}} \sqrt[3]{\frac{4\pi}{3 \times N_{\text{TRISO}}}}
\]

where,

\( r_{\text{zone}} \) is radius of the fueled-zone of pebble.

Table 4 shows the characteristics of plutonium content with TRISO number and SC lattice pitch used in the calculation. PuO\(_2\) kernel concentration is calculated by a simple formula and presented in table 5. The concentration of the coating covered the kernel is calculated by more simple formula and presented in same table. The concentrations of graphite matrix and graphite shell which is identical are presented in table 6. Figure 1 illustrates the MCNPX model of TRISO particle in fuel pebble.

**Table 4.** Characteristics of plutonium content.

| Pu content / pebble (g) | Number of TRISO | Lattice pitch SC (cm) |
|-------------------------|-----------------|-----------------------|
| 0.50                    | 7526            | 0.20567               |
| 0.75                    | 11,288          | 0.17967               |
| 1.00                    | 15,051          | 0.16324               |
| 1.25                    | 18,814          | 0.15154               |
| 1.50                    | 22,577          | 0.14261               |

**Table 5.** Concentration of TRISO coated particle [10].

| Kernel PuO\(_2\)         | Buffer         | iPyC/oPyC          |
|--------------------------|----------------|--------------------|
| \(^{239}\)Pu              | 6.01178x10\(^{-4}\) | \(^{242}\)Pu 1.54539x10\(^{-3}\) |
| \(^{239}\)Pu              | 1.24470x10\(^{-2}\) | \(^{16}\)O 4.60983x10\(^{-2}\) |
| \(^{240}\)Pu              | 5.44599x10\(^{-3}\) | \(^{10}\)B 1.14694x10\(^{-7}\) |
| \(^{241}\)Pu              | 3.00965x10\(^{-3}\) | \(^{11}\)B 4.64570x10\(^{-7}\) |

| Coating layers          | \(^{12}\)C 5.26449x10\(^{-2}\) | \(^{12}\)C 9.52621x10\(^{-2}\) |
| SiC                     | 4.40158x10\(^{-2}\) | \(^{10}\)Si 1.47944x10\(^{-2}\) |
| SiC                     | 2.22871x10\(^{-2}\) | \(^{12}\)C 4.77240x10\(^{-2}\) |

**Table 6.** Concentration of graphite matrix and graphite shell [10].

| Graphite matrix | Graphite shell |
|-----------------|---------------|
| \(^{12}\)C 8.77414x10\(^{-2}\) | \(^{12}\)C 8.77414x10\(^{-2}\) |
| \(^{10}\)B 9.64977x10\(^{-9}\) | \(^{10}\)B 9.64977x10\(^{-9}\) |
| \(^{11}\)B 3.90864x10\(^{-8}\) | \(^{11}\)B 3.90864x10\(^{-8}\) |
3.2. Fuel Pebble Model

The randomness of fuel pebbles in the reactor core was modeled with body-centered cubic (BCC) lattice. There are two fuel pebbles in the BCC lattice: 1 is placed in the center and 1/8 in each corner of the cube. The BCC cubic lattice was defined as UNIVERSE. Similar to the modeling of fuel pebble, the reactor core were then modeled by applying LATTICE and FILL options with repetitive structures. The lattice pitch was calculated to be 7.185259 cm to adjust 359,548 fuel pebbles occupying the reactor core with packing fraction of 0.61.

Figure 1. The MCNPX model for TRISO particle in fuel pebble.

Figure 2. The MCNPX model of fuel pebble in the reactor core.
Moderator pebble in the conical region was also modeled with the same lattice and packing fraction with the fuel pebble. The edge effect of associated to the use of a repeated structure in fuel pebble modeling can be ignored because the packing fraction for the TRISO particles inside the fueled-zone of the pebble is very low, around 1.73-5.19%. However, the same edge in core modeling effects can lead to non-negligible conditions in reactor calculation. Therefore, the packing fraction of 0.61 is reduced to 0.59 as a correction to eliminate the truncated pebble contribution in the surface of the core wall. Modeling of the reactor structure such as reflector, cooling channels, control rods and carbon layers around the reactor can be addressed in not difficult way. Modeling procedure of pebble bed reactor in detail and comprehensively with MCNP can also be found in our previous studies [11,12]. The CINDER90 modul integrated in the MCNPX was used to perform the nuclide inventory calculations as a function of fuel burnup. Figure 2 illustrates the MCNPX model of fuel pebble in the reactor core.

4. Result and Discussion
Temperature coefficient of reactivity is the response of the neutronic system to variations in temperature and defined as the ratio of reactivity change and temperature change. In this study, the calculated temperature coefficient of reactivity is a summation over the temperature coefficient of fuel, moderator and reflector. All calculations were done at various temperatures identified in ENDF/B-VII library. The problem was run using 110 cycles including 10 cycles skipped of 5000 particles per cycle to obtain exact results. The thermal scattering library $S(\alpha, \beta)$ which depends on temperature was applied to consider the interaction of thermal neutrons with graphite moderator, graphite reflector, and graphite contained in the entire fuel pebble under the energy of ~4 eV. Initial neutron source was placed in the fuel kernel to lead fission reactions throughout the reactor system. Vacuum conditions was done on the outer boundary of pebble bed reactor.

Figure 3. Reactor multiplication factor ($k_{\text{eff}}$) as a function of fuel burnup.

Figure 4. Temperature reactivity coefficient of 0.50 g Pu/pebble.
The calculation result of the reactor multiplication factor $k_{\text{eff}}$ for the case of plutonium loading varied from 0.5 g to 1.5 g per fuel pebble is illustrated in figure 3. It can be observed here, the $k_{\text{eff}}$ value of 1.25 g Pu/pebble at the beginning of the operating cycle is less than those of 1.50 g Pu/pebble. The $k_{\text{eff}}$ initial value of 1.00 g Pu/pebble is smaller than those of 1.25 g Pu/pebble. And so on, the $k_{\text{eff}}$ initial value of 0.50 g Pu/pebble is smaller than those of 0.75 g Pu/pebble. This phenomenon occurs because when the plutonium content per pebble is low, the presence of abundant graphite causes the better neutron thermalization. Consequently, the neutron flux will change to be higher and more fuel is burned. That is why the $k_{\text{eff}}$ curve of 0.5 g Pu/pebble declines sharply with the increase in fuel burnup. The greater Pu loading per pebble yields $k_{\text{eff}}$ curve declines slighter. The fuel with high Pu content per pebble may reach long burnup cycle. However, in general almost all the fuel can achieve burnup more than 450 GWd/T.

![Figure 5. Temperature reactivity coefficient of 0.75 g Pu/pebble.](image1.png)

![Figure 6. Temperature reactivity coefficient of 1.00 g Pu/pebble.](image2.png)

The calculation results of temperature reactivity coefficient are illustrated in figures 4 to 8. In the case of 0.5 g to 1.25 g plutonium loading per fuel pebble, each temperature coefficient of reactivity shows a similar tendency. These fuels become more negative with increasing temperature in fresh condition and burnup 200 and 400 GWd/T. Temperature reactivity coefficient values are in the range of $-5.0 \times 10^{-6}$ and $-1.5 \times 10^{-4} \Delta k/\text{kJ} \degree \text{C}$ for the temperature range of 50 °C and 1000 °C. The fuel with burnup 600 GWd/T shows the most negative temperature coefficient of reactivity at low temperature, but then reducing drastically and become positive at temperature greater than 700 °C. This means that the reactor containing 0.5 g-1.25 g Pu/pebble at high burnup has less favorable safety features if it operated at high temperature.
For the case of 1.5 g plutonium loading per fuel pebble, temperature coefficient reactivity shows a different trend. Fuel with burnup of 200 GWD/T shows more negative temperature coefficient of reactivity at temperatures of less than 600 °C, but turn out to be less negative above 600 °C. The temperature coefficient value at temperature of 1000 °C is -2.0×10^{-5}Δk/k°C slightly more negative than -1.5×10^{-5}Δk/k°C at temperature of 150 °C. The fuel with burnup of 400 GWD/T shows less negative temperature coefficient of reactivity than those of 200 GWD/T, but both have similar characteristics. At temperature of greater than 950 °C, the temperature coefficient becomes positive. On the other hand, the fuel with burnup of 600 GWD/T shows the most negative temperature coefficient of reactivity at temperature of 150 °C, but later reducing and turning positive after temperature of 300 °C. This temperature coefficient returns to negative value at temperature above 900 °C. These results give an impression that the use of fuel with high Pu content per pebble at high burnup should be considered carefully from the core safety aspect because it could affect transient behavior into a fatal accident situation.

Figure 7. Temperature reactivity coefficient of 1.25 g Pu/pebble.

Figure 8. Temperature reactivity coefficient of 1.50 g Pu/pebble.

5. Conclusion
Study on characteristic of temperature coefficient of reactivity for plutonium core of pebble bed reactor has been conducted. A series of calculations with plutonium loading varied from 0.5 g to 1.5 g per fuel pebble were performed by the MCNPX code and ENDF/B-VII library. The calculation results show that the $k_{\text{eff}}$ curve of 0.5 g Pu/pebble declines sharply with the increase in fuel burnup while the greater Pu loading per pebble yields $k_{\text{eff}}$ curve declines slighter. The fuel with high Pu content per pebble may reach long burnup cycle. From the temperature coefficient point of view, it is concluded that the reactor containing 0.5 g-1.25 g Pu/pebble at high burnup has less favorable safety features if it is operated at high temperature. The use of fuel with Pu content of 1.5 g/pebble at high burnup should be considered carefully from core safety aspect because it could affect transient behavior into a fatal accident situation.
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