The Effects of U-233 Impurity on U-232 and Tl-208 Buildup in Experimental Power Reactor with Thorium-Based Fuel

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Abstract. The existence of Tl-208 in thorium fuel cycle is a double-edged sword. Tl-208 is a high-energy 2.6 MeV gamma emitter, which acts as an effective proliferation barrier while simultaneously complicating the handling of the spent fuel. To ensure the safety of the latter, the buildup of both Tl-208 and its parent, U-232, are necessary to be understood. This paper attempts to analyse the buildup of U-232 and Tl-208 in the Reaktor Daya Eksperimental (Experimental Power Reactor/RDE) fuel based on thorium cycle, using various U-233 isotopic vectors. The simulation result shows that U-232-contaminated fresh fuels ended up with higher Tl-208 and U-232 activities at the end of cycle (EOC) compared with uncontaminated fresh fuel. However, their U-232 build-up rate are lower and even negative at one case. Then, lower U-233 purity caused a higher U-232 and Tl-208 activities at EOC. This result implies a considerable difference of isotope buildup between the various U-233 vectors. Consequently, the thorium cycle-based RDE spent fuel handling should consider the isotopic vector of U-233 used in fresh fuel.

Keywords: RDE, thorium, Tl-208, U-232, ORIGEN2.1

1. Introduction
Thorium fuel cycle possesses an advantage in term of anti-proliferation, that is U-232 existence. It is formed as the fuel is being irradiated and quickly decays into Tl-208, a high-energy 2.6 MeV gamma radiation emitter. This high-energy radiation complicates the effort of fuel diversion into nuclear weapon [1,2]. On the other hand, U-232 existence also makes spent fuel handling more difficult. The very same gamma release ensures the necessity of specialised fuel handling system. The requirement of radiation shielding in the spent fuel canister could also possibly be thicker [3,4].

Pure thorium cycle uses U-233 as the fissile isotope. Typically, researches concerning thorium fuel cycle utilisation assume that the U-233 is pure [5,6]. This is not currently realistic, since U-233 obtained from thorium irradiation always contain impurities in form of other uranium isotopes, including U-232 [7]. When irradiated, contaminated U-233 could yield U-232 in different concentration with pure U-233.

Experimental Power Reactor (Reaktor Daya Komersial/RDE) is an experimental-scale nuclear power reactor designed by Indonesian National Nuclear Energy Agency (BATAN). It employs low-enriched uranium (LEU) as the fuel. Nonetheless, RDE can possibly use other fuel cycle, including...
Thorium fuel cycle. In such scenario, the effect of U-232 contamination in U-232 in U-233 concerning U-232 and Tl-208 buildup needs to be understood, so that the spent fuel handling can be performed safely.

This study analysed the pattern of Tl-208 and U-232 buildup in the thorium-based RDE fuel. Buildup and decay simulations were performed using ORIGEN2.1 computer code with HTGR-type reactor neutron cross-section library. Fissile U-233 is simulated in several isotopic vectors with various level of purity.

2. Theory

RDE adopted a pebble bed-core high temperature gas-cooled reactor (HTGR) as its design, with a thermal power of 10 MW. The fuel is in spherical form, called pebble, filled by thousands of TRISO-coated micro-sized LEU kernels [8–10]. In this study, LEU is replaced by thorium and U-233.

The main anti-proliferation characteristics in the thorium fuel cycle is its extremely low transuranic elements production and the existence of U-232 as a contaminant. U-232 is produced in a two-step reaction as explained in Equations 1 and 2.

\[
\frac{\text{n} + {}^{232}\text{Th} \rightarrow {}^{233}\text{Th}}{22 \text{ min}} \rightarrow {}^{233}\text{Pa} + e^- \rightarrow {}^{233}\text{U} + e^- \]

(1)

\[
\frac{\text{n} + {}^{232}\text{U} \rightarrow 2\text{n} + {}^{232}\text{U}}{27 \text{ d}} \rightarrow {}^{231}\text{Pa} + e^- 
\]

(2)

\[
\frac{\text{n} + {}^{231}\text{Pa} \rightarrow {}^{232}\text{Pa} \rightarrow {}^{232}\text{U} + e^-}{1.3 \text{ d}}
\]

Reaction (1) predominantly occurs during the beginning of fuel irradiation. After Pa-231 accumulates, reaction (2) took over as the main contributor of U-232 formation [2]. The energy threshold of (n,2n) reaction is around 6 MeV. Therefore, U-232 production depends on fast neutron flux [11]. This could mean that reactor that operates in epithermal spectrum or reduced-moderation may generate more U-232 compared to very thermalised reactor.

ORIGEN2.1 computer code was employed to simulate the buildup and decay of U-232 and Tl-208. The code is known to be highly reliable in calculating radioactivity buildup and decay. ORIGEN2.1 works using Bateman Equation and one group-averaged neutron cross-section library. The former is represented in the following differential equation [12,13].

\[
\frac{dX_i}{dt} = \sum_{j=1}^{N} l_{ij} \tilde{A}_j X_j + \varphi \sum_{k=1}^{N} f_{ik} \sigma_k X_k - (\lambda_i + \varphi \sigma_i + r_i)X_i + F_i, \quad i = 1, ..., N
\]

(3)

where \( X_i \) denotes the density of nuclide \( i \), \( N \) is the nuclides number, \( l_{ij} \) is radioactive disintegration fraction by other nuclide that leads to formation of \( i \), \( j \) is the iterations number from \( j = 1 \) to \( j = N \), \( \varphi \) is position and energy-averaged neutron flux, \( f_{ik} \) is the neutron fraction absorbed by other nuclides that leads to formation of species \( i \), \( \sigma_k \) is the averaged neutron absorption cross section of nuclide \( k \), \( r_i \) is the continuous removal rate of nuclide \( i \), and lastly \( F_i \) is the continuous feed rate of nuclide \( i \).

ORIGEN2.1 requires a reactor-specific neutron cross-section library to be able to generate nuclide data properly. This study employed RDE.LIB cross-section library. The library has been verified to be suitable to calculate radionuclide inventory of HTGR-type reactor, including RDE [8,14].

Previous researches regarding radionuclide inventory on various type of HTGR have been performed [8,12,15] including thorium-based fuel [16]. However, the latter was performed only for one isotopic vector of U-233, also it did not specifically discuss Tl-208 and U-232 buildup.

3. Methodology
Buildup and decay simulation of Tl-208 and U-232 was performed using ORIGEN2.1 code with RDE.LIB cross-section library. ORIGEN2.1 calculate the buildup and decay of radionuclides as a point reactor. Consequently, in this case, it is not suitable to simulate the full core. Instead, the calculation was performed as one pebble.

There are two nuclide depletion schemes available in ORIGEN2.1, namely constant power (IRP) and constant flux (IRF) [17]. The latter is used if time-dependent neutron flux data is available. Since the data is currently unavailable, IRP scheme was used instead. The power was set at constant value of 3.7 kW/pebble, assuming equilibrium core [12].

The fuel loading of thorium-fuelled HTGR have been discussed previously [5,18] but the latter did not provide optimum thorium fuel composition for use in HTGR. Therefore, fuel loading mentioned in [5] will be used as the reference fuel.

It is also currently unknown how long the effective full-power days (EFPD) of RDE using multi-pass thorium cycle. Therefore, this study assumed that the EFPD is the same with LEU-cycle RDE fuel, that is 1080 days. Future works should consider the full EFPD for thorium-fuelled RDE to yield better representation. The waiting time before fuel recirculation was set at 40 days.

Thorium-cycle RDE fuel parameters are shown in Table 1.

| Parameter                  | Value       |
|----------------------------|-------------|
| Pebble thermal power       | 3.7 kW      |
| Fuel type                  | (U,Th)O₂    |
| U-233 enrichment           | 8%          |
| Heavy metal loading        | 6 g         |
| Pebble mass                | 200 g       |
| Fuel loading scheme        | Multi-pass  |
| EFPD                       | 1080 days   |

Isotopic vector of U-233 used in this study is varied into pure and contaminated vectors. Several references are available for contaminated U-233 isotopic vector [7,19–21]. Thus, seven variation of isotopic vector will be calculated; one pure and six contaminated. The vectors are provided in Table 2.

| Isotope | Vector 1 | Vector 2 | Vector 3 | Vector 4 | Vector 5 | Vector 6 | Vector 7 |
|---------|----------|----------|----------|----------|----------|----------|----------|
| U232    | 0%       | 0.04%    | 0.35%    | 0.05%    | 0.00%    | 0.00%    | 0.00%    |
| U233    | 100%     | 85.99%   | 66.74%   | 63.03%   | 92.80%   | 85.20%   | 79.40%   |
| U234    | 0%       | 11.81%   | 20.97%   | 25.12%   | 6.50%    | 12.50%   | 16.70%   |
| U235    | 0%       | 1.68%    | 6.24%    | 5.92%    | 0.65%    | 2.00%    | 3.40%    |
| U236    | 0%       | 0.48%    | 5.66%    | 5.88%    | 0.04%    | 0.20%    | 0.50%    |
| U238    | 0%       | 0.00%    | 0.04%    | 0.00%    | 0.00%    | 0.00%    | 0.00%    |

4. Result and Discussions
The calculation result is divided into two categories, U-232 Buildup and Tl-208 Buildup. The analysis was focused on radioactivity on both isotopes and gamma release exclusively for Tl-208.

4.1. U-232 Radioactivity
Among the simulated U-233 vectors, three of them have already contaminated with U-232 in the beginning of cycle (BOC). Supposedly, their U-232 activity should be the highest at the end of cycle (EOC). This was proven to be true, but not in the expected pattern, as shown in Figure 1.
Vector 3 contains the highest amount of U-232 at the BOC. However, its radioactivity was decreasing over time instead of increasing, amounting for total of 29.15% radioactivity decrease. Such pattern indicates the existence of “saturation point,” that is when the U-232 contamination reached certain concentration so that neutron capture rate is higher than buildup rate. This saturation point may differ each other depending on many factors, which will be discussed later.

Vector 2 and 4 show different pattern. Their U-232 activities at the beginning of irradiation were also decreased, but after reaching certain level of radioactivity, they started to accumulate. This phenomenon also shows a saturation point, despite this being the bottom saturation instead of top saturation. U-232 radioactivity in the aforementioned vectors were higher at EOC than BOC, indicating that buildup was more dominant than burning.

Bottom saturation point does not seem to have an exact value. This is due to the fact that the buildup turning point was different for each vector. The bottom saturation points of vector 2 and 4 were 3.692x10^{-3} Ci and 4.851x10^{-3} Ci, respectively. Such difference is directly influenced by the contaminant vector itself. The higher the level of contamination, the less fission from U-233 and more neutrons are absorbed by Th-232, increasing the possibility of (n,2n) reaction. This is shown by the radioactivity of Pa-231 in vector 4 is higher than vector for both loadings, as shown in Table 3.

**Table 3. Pa-231 radioactivity on vector 2 dan 4 at EOC**

| Isotopic vector | Radioactivity (Ci) |
|----------------|--------------------|
| 2              | 1.16x10^{-5}       |
| 4              | 1.28x10^{-5}       |

Although vector 1, 5, 6, and 7 were not contaminated with U-232 at BOC, their U-232 buildup rate were different. Vector 1, which assumed to be completely pure from any contamination, has the lowest U-232 buildup rate. The activity recorded at EOC was 2.091x10^{-3} Ci. Meanwhile, vector 7 with highest contaminant, recorded U-232 activity at EOC as large as 2.663x10^{-3} Ci.

Similar with the case of vector 2 and 4, this difference was caused by impurity level of U-233 isotope. Less U-233 content due to impurities led to more neutron absorption by thorium, which in...
return increasing (n,2n) reaction rate and subsequently more U-232 formation. Activities of Pa-231, the result from (n,2n) reaction, were consistently higher in lower U-233 purity vectors, as shown in Table 4.

**Table 4. Pa-231 radioactivity on vector 1, 5, 6, dan 7 at EOC**

| Isotopic vector | Radioactivity (Ci) |
|-----------------|--------------------|
| 1               | 1.082x10^{-5}      |
| 5               | 1.121x10^{-5}      |
| 6               | 1.159x10^{-5}      |
| 7               | 1.186x10^{-5}      |

**4.2. TI-208 Radioactivity and Gamma Release**

TI-208 buildup heavily depends on U-232 concentration at the BOC. In reality, U-232 contaminated vectors should already contain TI-208 at the BOC, since U-232 constantly decays. There is no data to estimate the initial activity of TI-208 and it would be very complicated to do so. Therefore, this study ignored TI-208 activity at the BOC for all vectors. The buildup of TI-208 is shown in Figure 2.

![TI-208 buildup chart](image)

**Figure 2.** TI-208 buildup over time for various U-233 isotopic vector.

It is quite obvious that vector 3 has the fastest TI-208 buildup rate, bearing in mind that its U-232 contamination at the BOC is the highest. Generally, the TI-208 buildup was more drastic in the first 500 days before starting to flatten. This especially visible after day 884, only increasing during the waiting time outside the core.

The significantly higher activity on vector 1 dwarfed the buildup chart of other vectors. Better perspective by omitting vector 3 is provided in Figure 3.
Figure 3. Tl-208 buildup over time omitting vector 3

Vector 2 and 4 encountered Tl-208 buildup relatively fast, although not as fast as vector 3. The rest of the vectors have the lowest buildup rate, thanks to the absence of U-232 at the BOC. In line with U-232 case, vector 1 has the slowest Tl-208 buildup rate due to its Th-232 neutron capture being the lowest. The buildup of Tl-208 was proportional to U-232 buildup rate.

Assessment of gamma release is crucial to determine proper fuel handling after being irradiated in the reactor core. Therefore, decay calculation of spent fuel after discharge is performed until 100 years after EOC. Gamma release value at EOC is provided in Table 5.

| Isotopic vector | Gamma release rate (photons/s) |
|-----------------|--------------------------------|
| 1               | 6.96x10^6                     |
| 2               | 2.89x10^7                     |
| 3               | 1.91x10^8                     |
| 4               | 3.65x10^7                     |
| 5               | 7.66x10^6                     |
| 6               | 8.43x10^6                     |
| 7               | 9.04x10^6                     |

Gamma release of vector 1 was recorded at only 3.65% of vector 3, indicating a non-negligible difference. Meanwhile, gamma release difference from vector 1 with vector 5, 6, and 7 were recorded at 9.13%, 17.43%, and 22.99%, respectively. Those values are approximately similar with their respective level of isotopic impurity. Thus, it can be safely said that the increase of gamma release rate of Tl-208 is proportional to the increase of U-233 impurity level.

Tl-208 decay pattern, as well as most of U-232 daughter products, is different with general radionuclides. Instead of decaying, Tl-208 is building up until year 10 after discharge, before starting to decay with slower rate compared with its buildup. Tl-208 decay after irradiation is shown in Figure 4.
Figure 4. Tl-208 buildup and decay after irradiation.

Gamma release of vector 3 at year 10 after discharge was recorded at $2.98 \times 10^8$ photons/s. Compared to vector 1, the release is 12.37 times higher. As a comparison, at discharge, vector 3 gamma release was 27.41 times higher than vector 1. Such decrease of gamma release comparison was also found relative to other vectors. This indicates that Tl-208 buildup rate in the first 10 years after discharge are higher in pure U-233 vector. Whether this pattern is important to safety of spent fuel handling or not depends on the assumptions beyond the scope of this study.

Tl-208 decay after year 10 showed lower decay rate for all vectors in both fuel loadings. For contaminated vectors, gamma release rates were lower in year 100 compared to year 0, whilst the opposite applied to uncontaminated vectors. Whether or not this pattern means that the contaminated vectors will end up with lower Tl-208 gamma release in longer term cannot be concluded just yet.

5. Conclusion
U-232 formation in thorium-based RDE fuel encounters a saturation point, so that when U-232 contamination reached a certain activity, its buildup rate becomes negative. On the opposite, after its activity decreased until a certain point, U-232 starts to buildup once again. This pattern affects the radioactivity and gamma release of Tl-208, especially for the vectors contaminated by U-232 at BOC. Meanwhile, vectors uncontaminated by U-232 at BOC have its U-232 and Tl-208 buildup rate proportional to the contamination level. The difference of U-232 and Tl-208 buildup between pure and contaminated vectors, especially vectors contaminated by U-232, relatively large and necessitates different spent fuel handling mechanism. Future works should calculate their activity and gamma release at full EFPD of thorium cycle RDE, which data is unavailable at the moment.

Acknowledgments

This work was funded by research budget of Centre for Nuclear Reactor Technology and Safety, National Nuclear Energy Agency, Fiscal Year 2019 and research grant from Ministry of Research, Technology and Higher Education of Indonesia.

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