Analysis of Burnable Poison Effect on Combined (Th-U)O\textsubscript{2} Fuel Cycle Performance in 800 MWt Long-Life PWR

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Abstract

Thorium based fuel especially on thermal reactor has been a great future sight. To produce electricity, thorium should be combined by any fissile material. Many research has been conducted to examine feasibility of thorium fuel. In this present study, burnable poison effect on combined thorium-uranium based fuel for long life PWR at thermal power output 800 MWt has been conducted. The fuel in this study consists of ThO\textsubscript{2} combined by 40 – 60% enriched UO\textsubscript{2} in an acceptable enrichment (13-20 % U-235). To reach long life operation time, fuel volume fraction is set to be 40-60 %. Burnable poison on this design includes for 0.005-0.06 % Gd\textsubscript{2}O\textsubscript{3}, 0.5-2 % Pa-231, and 0.5-2 % Np-237. Gd\textsubscript{2}O\textsubscript{3} has very high thermal neutron absorption cross section, small amount of Gd\textsubscript{2}O\textsubscript{3} relatively sufficient to reduce excess reactivity, but the effect of this material effective work in early burnup period. Np-237 has 190 barns thermal neutron capture cross section which can be used to minimize excess reactivity. Pa-231 has higher thermal neutron capture cross section (280 barns) rather than Np-237 (190 barns), hence the effect of Pa-231 stronger than Np-237 to minimize excess reactivity. In the other side, Pa-231 could compensate fissile isotope in the further burnup step. The optimized design of this study reached 10 years operation time with excess reactivity smaller than 2.5% \(\Delta k/k\).

Keywords: Burnable Poison, Long-Life PWR, Thorium, Uranium

1. Introduction

Energy is one of the most valuable resource for all creature in the world. The world energy demand may increase up to 47% by 2035 if energy policies in this current age still implemented until next 2035, according to the International Energy Agency (IEA)\textsuperscript{1}. Nuclear resource is one of the significant energy resources. Nuclear power has been operated for more than 60 years in the world. To meet energy demand, some country has accepted contribution of nuclear energy as a safe and economic energy resource which has approximately 439 operated nuclear reactor power plant in the world\textsuperscript{2}. The most widely used nuclear reactor is Pressurized Water Reactor (PWR) to produce electricity. Developing countries, contribute 74% of the increase global energy demand to compensate fast growing in economies and populations\textsuperscript{3}.

Small nuclear reactor would be an some important energy resource for developing countries to be used in many remote area promote nuclear energy. Small Modular Reactor (SMR) research have been conducted since long time ago to increase reactor operation and safety performance. Small long life pressurized water reactor is one of adopted SMR for near future usage. Research about small long life pressurized water reactor has been conducted for many types. Thorium based fuel has been implemented to increase operation time since this isotope has several advantage rather than conventional fuel.

The abundant of thorium three times greater than uranium in a natural resource, this advantage could provide long term supply of nuclear fuel. Th-232 is a fertile isotope which can be converted to fissile isotope U-233. Capture cross section of thorium is three times greater than uranium, this parameter make the reactor has higher
conversion ratio which can provide longer operation time. If several neutron captured by thorium, it will produce fissile nuclide U-233 which has higher thermal fission cross section than U-235. The advantage of this parameter could consume smaller percentage of U-233 in a fuel. If thorium based fuel implemented in a nuclear reactor, plutonium portion in the spent fuel could be reduced. Another perspective, ThO$_2$ also has 50% higher thermal conductivity compared to that of UO$_2$.

Taking into consideration Th-232 as a fertile isotope, it is needed to add fissile isotope. Research has been conducted to insert fissile isotope as known as U-233, U-235 or Pu-239 as in the fuel.

In a previous paper, we have studied about feasibility of thorium combined by 30-40% uranium (10-15% U-235 in a uranium fuel component) fuel and implemented gadolinium as a burnable poison at thermal power 600-1000 MWt. The result from this study has reactivity around 10% (maximum $k_{eff}1.10$). In the present study, we extended our study on feasibility of burnable poison such as protactinium, neptunium, and gadolinium to reduce excess reactivity but still provide longer reactor operation time at thermal power 800 MWt. This study has implemented combination (Th-U)O$_2$ as fuel with 40-60% UO$_2$ with acceptable enrichment. The purpose of this design study is to reach long life pressurized water reactor with 10 years minimum operation time and small excess reactivity maximum (3% $d_{k}/k$).

2. Theoretical Background

Nuclear reactor analysis has many aspect to be examined. In this paper, we studied about neutronic performance of the reactor core. The neutronic analysis includes transport equation to analyze neutron behavior in the solution of reactor core. Transport equation relatively hard to be solved in analytical and computational solution. The simpler equation to be solved is multigroup diffusion equation which has four term. In a steady state condition, the diffusion equation can be written as.

\[
-D_x \frac{\partial}{\partial x} \phi_x + \sum_{g \neq x} S_{g,x} \phi_g - \frac{\phi_x}{k_{eff}} \sum_g \sum_{x' \neq x} S_{g,x'} \phi_{x'} + \sum_{x'' \neq x} \phi_{x''} = 0
\]  

\[
(1)
\]

- $f_g$: Neutron flux.
- $k_{eff}$: Effective multiplication factor.

The equation can analyze neutron flux distribution in the reactor core and evaluate neutron multiplication symbolized by $k_{eff}$. Multiplication factor is factor defined as number of neutron in one generation divided to number of neutron in preceding generation.

\[
k = \frac{\text{number of neutron in one generation}}{\text{number of neutron in preceding generation}}
\]

This parameter determine nuclear reactor condition, $k = 1$ critical, $k > 1$ supercritical, $k < 1$ subcritical.

Neutron flux from fission reaction in a reactor core depends on core geometry, and composition. Power distribution profile can be described from flux behavior obtained from diffusion equation. The power distribution profile formulated with the following

\[
q^{n}(r) = \sum_{i} w_{i}^{n} N_{i}^{n}(r) \int_{0}^{\infty} dE \sigma_{f}^{i}(E) \phi(r, E)
\]

\[
(3)
\]

- $w_i$: Energy produced per fission reaction.
- $N_i$: Atomic density for isotope $i$.
- $\sigma_{f}^{i}$: Microscopic fission cross section for isotope $i$.
- $f(r,E)$: Neutron flux with dependence of position and energy.

3. Reactor Design Parameter

3.1 Fuel Pin

In this study, reactor fuel which implemented is a combination ThO$_2$ and UO$_2$ mixed in single fuel pin. Percentage of UO$_2$ varied in 40-60% and the UO$_2$ enriched with 13-20% U-235 (percentage in whole fuel 5.4-12% U-235). This percentage of U-235 relatively higher than conventional PWR to compensate higher capture cross section of Th-232 than U-238. In burnup process, U-233 which has strong thermal fission factor will be produced and make an additional fuel while percentage of U-235 decreases.

Fuel fraction is also a significant factor to develop long life thermal nuclear reactor. Higher fuel fraction implicate to lower moderator fraction and decrease thermal neutron utilization$^{16}$. In this study, fuel fraction varied to 40-60%. Fuel pin arranged to 1.26 cm square cell pitch geometry with cylindrical fuel rod covered by 0.057 cm
zircalloy IV cladding. The radius of fuel rod pin depends on fuel volume fraction. The scheme of fuel pin is given in Figure 1.

3.2 Burnable Poison

To obtain long life reactor operation time, various burnable poison has been inserted as an additional composition in fuel rod. Gd$_2$O$_3$, Pa-231, and Np-237 has been studied as burnable poison to examine its effect in reactor operation time and reactivity. Burnable poison utilization can maintain long life operation and decrease reactivity the burnable poison isotope has thermal neutron absorption cross section more than 1000 times higher than thorium and uranium.

3.2.1 Gadolinium

Gadolinium has very high thermal absorption cross section. In the nature, of six stable isotope of gadolinium as a mixture, Gd-154 (2.18%), Gd-155 (14.8%), Gd-157 (15.67%), Gd-156 (20.47%), Gd-160 (21.86%), and Gd-158 (24.84%) and one radioactive isotope Gd-152 (0.2%). Gd-157 has highest nuclear absorption cross section, about 259,000 barns. Mixture of all isotope in natural gadolinium absorption cross section is around 49,000 barns$^{11}$. In this study, small amount of Gd$_2$O$_3$ in mixed fuel is examined to reduce initial reactivity. We have evaluated 0.01-0.06 % Gd$_2$O$_3$ as a composition of reactor fuel.

3.2.2 Neptunium-237

Neptunium has been produced in conventional reactor by decay product of plutonium which obtained from U-238. The stable isotope is Np-237 has half-life of 2,144,000 years$^{11}$. Capture cross section of Np-237 around 180 barns$^{12}$. In this study, 0.5-2 % Np-237 inserted as fuel composition to evaluate reactor criticality.

3.2.3 Protactinium-231

Protactinium (Pa) which has atomic number 91 is the rare nuclide, rarer than radium. This could be found from uranium ores with amount 0.34 ppm uranium. Pa-231 is the most valued isotope of Pa which has long-lived existence with half-life of 32,760 years$^{11}$. Pa-231 has high thermal neutron capture cross section, around 293 barns$^{13}$. Similar to Np-237, 0.5-2 % Pa-231 inserted as fuel composition to evaluate reactor criticality.

3.4 Reactor Core

The reactor core design parameter arranged to thermal power output 800 MWt with power density $\sim$43 W/cc. Thus the reactor dimension is arranged to obtain this value, and reactor core dimension to suitable this value is 276.8 cm diameter and 307.44 cm height. To decrease neutron leakage, active core is covered by 22.68 cm reflector width with stainless steel as a reflector material.

To examine neutronic behavior in reactor core, of reactor core is in cylindrical R-Z geometry and divided into 3 region in radial direction. Every region has similar width. Summary of all parameter included in this study given in Table 1.

4. Calculation Method

Calculation of this study carried out by SRAC Code System and library data from SRACLIB-JDL32. To examine neutronic behavior on nuclear reactor core, first of all, macroscopic cross section data of every isotope should be calculated. To calculate macroscopic cross section data, fuel pin calculation should be calculated first. There is PIJ module to calculate this pin calculation in SRAC module. PIJ calculation can calculate several parameter include multiplication factor, conversion ratio, fission product, burnup level, neutron flux, microscopic and macroscopic cross section data, etc. Data of cell calculation saved in user library data. Core calculation can be calculated after macroscopic cross section data triggered. CITATION module can be used to core calculation to found out effective multiplication factor ($k_{\text{eff}}$), average power density, power distribution in axial and radial direction$^{14}$. Reactor core geometry in CITATION module adopted 2D cylindrical
R-Z geometry divided into 3 region in radial direction. The shape of reactor core geometry shown in Figure 2.

<table>
<thead>
<tr>
<th>Parameter</th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>Active core diameter</td>
<td>277.2</td>
</tr>
<tr>
<td>Active core height</td>
<td>307.44</td>
</tr>
<tr>
<td>Power density</td>
<td>43.12</td>
</tr>
<tr>
<td>Reflector width</td>
<td>22.68</td>
</tr>
<tr>
<td>Reflector material</td>
<td>Stainless steel + H$_2$O</td>
</tr>
<tr>
<td>Fuel (Th-U)$_2$O$_2$UO$_2$</td>
<td></td>
</tr>
<tr>
<td>UO$_2$ percentage</td>
<td>40 - 60 %</td>
</tr>
<tr>
<td>U-235 enrichment</td>
<td>13 - 20 %</td>
</tr>
<tr>
<td>Burnable poison</td>
<td>0.01 - 0.05 % Gd$_2$O$_3$</td>
</tr>
<tr>
<td></td>
<td>0.5 - 2 % Pa-231</td>
</tr>
<tr>
<td></td>
<td>0.5 - 2 % Np-237</td>
</tr>
<tr>
<td>Pin pitch</td>
<td>1.26 cm</td>
</tr>
<tr>
<td>Fuel volume fraction</td>
<td>40 – 60%</td>
</tr>
<tr>
<td>Clad thickness</td>
<td>0.057 cm</td>
</tr>
<tr>
<td>Moderator</td>
<td>H$_2$O</td>
</tr>
</tbody>
</table>

Before discuss about the effect of burnable poison in this study, first of all, we should examine multiplication factor of reactor core without burnable poison. The result of this examination shown in Figure 3 for 40% UO$_2$ composition and Figure 4 is for 60% UO$_2$ composition.

Alphabet symbol A-F symbolized enrichment of uranium composition in each core region.

- Case A : 13% region 1, 14% region 2, 15% region 3.
- Case B : 14% region 1, 15% region 2, 16% region 3.
- Case C : 15% region 1, 16% region 2, 17% region 3.
- Case D : 16% region 1, 17% region 2, 18% region 3.
- Case E : 17% region 1, 18% region 2, 19% region 3.
- Case F : 18% region 1, 19% region 2, 20% region 3.

According to Figure 3, multiplication factor increases if enrichment increases.

Higher percentage of uranium composition in a total fuel has increasing multiplication factor, but has not given much different in multiplication factor pattern. This is caused by increasing percentage UO$_2$ lead to increasing percentage of U-235 in a total fuel.

Meanwhile, higher fuel volume fraction impacted to lower multiplication factor. Pressurized water reactor is a thermal reactor which utilizing thermal neutron to generate fission reaction. In this type of reactor, moderator implemented to moderate fast neutron from fission reac-
tion to be slower neutron (thermal neutron), hence this neutron can be generate fission reaction. In higher fuel volume fraction, moderator fraction will be lowered and implicated to less thermal neutron amount to generate fission reaction.

Figure 5. Comparison between fuel without burnable poison at 40% and 60% UO$_2$, 55% fuel fraction.

Figure 6. Effect of fuel fraction, represented by moderator to fuel ratio (MFR/FF) in start-up.

In addition, higher fuel volume fraction impacted to lower moderator to fuel ratio (MFR, moderator volume fraction divided by fuel volume fraction). If MFR decrease, thermal utilization factor and resonance escape probability also impacted by this parameter. Figure 6 show that in a low MFR, multiplication factor also lower compared to higher MFR. According to reference, pattern of fig. 6 in under moderated condition. But if we look at pattern of 60% UO$_2$ curve, curve truncated at MFR = 1, this caused by when we evaluate the feasibility for 60% UO$_2$ at 40% fuel volume fraction (MFR = 1.25), the calculation was not converge. We analyzed that in this condition, the fuel is in over moderated condition. In practice, under moderated condition is a desirable condition for water-moderated reactor. In this condition, negative reactivity feedback will reach while fuel temperature increased$^{10}$.

5.2 Fuel with Burnable Poison Gd$_2$O$_3$

Evaluation of gadolinium utilization focused on beginning of life in reactor operation. It is as a result of gadolinium just consume neutron in an early time. Multiplication factor in early burnup step much lower rather than without gadolinium although with very low percentage in total fuel. It is caused by very high thermal neutron absorption cross section of gadolinium (~49,000 barns).

Figure 7. Fuel with 0.02% gadolinium, 40% UO$_2$, 18 – 20 enrichment U-235.

Figure 8. Fuel with 0.025% gadolinium, 60% UO$_2$, 18 – 20 enrichment U-235.

Figure 7 show that small amount of Gd$_2$O$_3$ as a burnable poison enough to lowering excess reactivity in early stage. But if we look at Figure 8, there is higher percentage of gadolinium in higher percentage of UO$_2$ composition. The effect of gadolinium much lowered rather than in lower percentage of UO$_2$.

In another point of view, higher fuel volume fraction make gadolinium effect decreased. The optimum feasibility study to utilize gadolinium as a burnable poison
shown in Figure 9. Reactivity still higher than 5% dk/k, is for 50-60 % UO₂, 16-20 % enrichment U-235 (8-12 % in total fuel) with 0.03% Gd₂O₃ at 60% fuel volume fraction.

Figure 9. Optimized fuel with Gd₂O₃ as burnable poison.

5.3 Fuel with Burnable Poison Np-237

Neptunium has been produced in conventional reactor by decay product of plutonium which obtained from U-238. This isotope has relatively high absorption cross section (~190 barns). As a nuclear waste, it is very useful to utilize as a burnable poison to reduce waste in the future and extend reactor operation life time with small reactivity. We have studied effect of this isotope as a burnable poison in this reactor design.

Figure 10 and Figure 11 shows effective multiplication factor from our study in this fuel type. Small amount of Np-237 (0.5-2 %) in total fuel has effected to decrease in multiplication factor. The pattern of kₚ curve relatively similar to fuel without burnable poison, but the reactivity of this fuel type relatively smaller than without burnable poison and has longer operation time for similar maximum kₚ. But the effect of this nuclide is not as significant as needed in our target. The lowest maximum reactivity from this fuel type still has high value, it is higher than 5% dk/k.

Figure 10. Fuel with Np-237, 45-50 % UO₂, 18-20 % enrichment U-235.

Figure 11. Fuel with Np-237, 60% UO₂, 18-20 % enrichment U-235.

5.4 Fuel with Burnable Poison Pa-231

Pa-231 has higher thermal capture cross section than Np-237, this condition will impacted to reach smaller reactivity than Np-237 as burnable poison in a similar amount of burnable poison in total fuel. In the other hand, Pa-231 would be an additional fuel in the next burnup step consider this isotope can be transmuted to U-233 via neutron capture reaction.

Figure 12 and Figure 13 show the effective multiplication factor for fuel type 40-45 % UO₂ and 60% UO₂, respectively. Pa-231 has shown more attractive result rather than previous two types of burnable poison. Reactivity from this result has shown smaller value and has longer operation time. It is caused by strong neutron absorption of Pa-231 which could decrease probability fissile isotope to interact with thermal neutron. Meanwhile, in the next stage burnup step, Pa-231 also produced fissile isotope U-233 as an additional fuel, so this fuel type could maintain longer operation time and has lower reactivity. This fuel type still has relatively wide gap reactivity in starting up and first burnup step. To decrease this gap, gadolinium could be a useful option to combine with Pa-231 to reach smaller maximum reactivity.

Figure 12. Fuel with Pa-231, 40-45 % UO₂, 18-20 % enrichment U-235.
5.5 Optimization Design

After some examination from this study, the optimized design has been reached. Figure 14 and Figure 15 show the optimized design for 45-55 % UO₂ and 60% UO₂, respectively.

In Figure 14, there is reached 10 years reactor operation time with reactivity < 3% dk/k. From previous discussion, combination of burnable poison gadolinium and Pa-231 has reached more attractive result than previous result. In Figure 15, 60% UO₂ composition could reached study target with 50-60 % fuel volume fraction. The lowest U-235 percentage in all fuel reached at 8% U-235 (50% UO₂, fuel case D) from total fuel, and maximum value reached at 12% U-235 (60% UO₂, fuel case F). Burnup level of this design around 40 GWd/t. Compared to conventional PWR, this value is in the range of burnup level of typical LWR (30-50 GWd/t)\(^1\).

To check power distribution of this optimized design, we will check for case 60% UO₂, 0.025% Gd₂O₃, 2% Pa-231, 55% fuel volume fraction, 18-20 % enrichment U-235.

Figure 16 show radial power distribution of examined fuel type. In a Beginning of Life (BOL), power distribution relatively flat with Power Peaking Factor (PPF) around 1.4. In End of Life (EOL), PPF of this fuel type 1.55. Peak of radial power shift from region 2 in BOL to region 1 in EOL.

In Figure 17, that show axial power distribution for this examined fuel type. PPF in axial direction around 1.45. Power distribution pattern in axial direction for BOL and EOL has a similar value because in axial direction has homogenous fuel.

There is 9 option to reach our purpose. In the future study, we can optimize this study with thermal hydraulic and safety analysis to get more comprehensive examination of this result.

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Figure 13. Fuel with Pa-231, 60% UO₂, 18-20 % enrichment U-235.

Figure 14. Optimization fuel at 45-55 % UO₂ composition.

Figure 15. Optimization fuel at 60% UO₂ composition.

Figure 16. Radial power distribution.

Figure 17. Axial power distribution.
6. Conclusion

Feasibility study of burnable poison on combined (Th-U)O₂ based fuel for long-life PWR has been conducted at thermal power output 800 MWt. Thorium as a fertile material has strong potential to be utilized as long life reactor fuel combined with any fissile material in thermal neutron spectrum. To reach long life operation time, it would be a great idea to insert burnable poison as a mixture of fuel composition. Gadolinium has strong absorption cross section, but the effect of this burnable poison type just in early stage of reactor operation. Np-237 as burnable poison also has potential to be a burnable poison since this isotope has capture cross section 190 barns. The most potential burnable poison is Pa-231 which has capture cross section around 280 barns and this isotope also can compensate fissile isotope in the next burnup step. To reach optimize lowest reactivity, there is an option to combine both Pa-231 and gadolinium as burnable poison. Optimize design of this study reach on 10 years operation time with reactivity smaller than 2.5% dk/k, this design reached on 45-60% UO₂, 40-60% fuel volume fraction 15-25% Gd₂O₃, and 1.5-2% Pa-231. The minimum percentage of U-235 in whole fuel is 8% and the maximum percentage at 12%. Burnup level of this study reached at ~40 GWd/t. The power peaking factor of this study in radial direction around 1.35 at BOL and 1.55 at EOL, and power peaking in axial direction around 1.45.

Thermal hydraulics and safety analysis will be conducted in future works. Reactor core dimension also will be rearranged to reach low power peaking factor.

7. References