Preliminary Study on Transmutation of Plutonium and Minor Actinides in Accelerator Driven System

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Abstract. As a free CO2 emission energy source, nuclear energy can supply the world electricity for very long time, even for thousand years. However, to increase the public acceptance, nuclear spent fuel (high level nuclear waste) should be manage to minimize its radio-toxicity. There are several methods of processing nuclear spent fuel including geological repositories, reprocessing it to be used as fuel again or doing transmutation. In this preliminary study, the nuclear high level waste is transmuted by using an Accelerator Driven Subcritical System (ADS). ADS is a system consisting of a subcritical reactor core and a particle accelerator. Nuclear waste which will be transmuted is plutonium (Pu) and minor actinides (MA). Pu and MA are mixed in the ADS fuel that consists of U-ThO$_2$(NO$_3$)$_2$, PuMA(NO$_3$)$_4$ dan HNO$_3$. Two type of plutonium vectors have been evaluated, those are reactor grade Pu (RGPu) and weapon grade Pu (WGPu). The ADS can maintain its sub-criticality condition for RGPuMA and WGPuMA with the concentration of PuMA-Nitrate are above 62.6 kmol/m$^3$ and 65.6 kmol/m$^3$, respectively.

Keywords: ADS, Plutonium, Minor Actinides, Spallation Reaction, Transmutation

1. Introduction

Energy is the key factor for supporting the economy growth. Most of the world energy sources come from fossil fuels that cause the environment deterioration due to the huge CO$_2$ emissions. Table 1 shows the energy consumption and the CO$_2$ emissions in the world from during 1990-2025 [1]. In 2015, the world electricity sources are: 66% of fossil fuels, 16% of hydro, 11 % of nuclear, 5% of renewable, and 2% of other sources [2]. The use of fossil fuels as main source of energy in the world will give greater contribution to global warming and climate change. A strategic solution is needed to overcome this problem, one of them by using nuclear energy that does not generate harmful gases. However, the use of nuclear energy until 2015 was less than 11% throughout the world.

The nuclear energy program has not been able to be accepted openly by the world community due to the following 3 main reasons: 1) Fear of proliferation, 2) Fear of accidents such as Chernobyl and Three Mile Island, and 3) Nuclear waste management issues [4-5]. However, the true issue regarding nuclear energy acceptance is the long-lived high level nuclear waste (HLW) management [6-7], since some of radioisotopes in HLW have more...
than million year of half-lived. HLW consist of plutonium (Pu), minor actinides (MA), and long-lived fission products (LLFP) [6-7].

Table 1. Energy consumption and CO₂ emissions in the world from year 1990-2025 [1]

| Region                | Energy consumption (× 10¹⁸ J) | CO₂ emissions (10⁶ metric tonnes carbon equivalent) |
|-----------------------|-------------------------------|--------------------------------------------------|
|                       | 1990  | 2001  | 2010  | 2025  | 1990  | 2001  | 2010  | 2025  |
| Industrialized countries | 192.9 | 223.1 | 253.3 | 304.2 | 2844  | 3179  | 3572  | 4346  |
| Developing countries  | 80.5  | 56.2  | 69.5  | 86.8  | 1337  | 856   | 1038  | 1267  |
| Asia                  | 55.4  | 89.7  | 116.2 | 184.2 | 1089  | 1640  | 2075  | 3263  |
| Middle East           | 13.8  | 21.9  | 26.6  | 38.0  | 231   | 354   | 420   | 601   |
| Africa                | 9.8   | 13.1  | 15.2  | 21.1  | 179   | 230   | 261   | 361   |
| Latin America         | 15.2  | 22.0  | 26.6  | 41.1  | 192   | 263   | 319   | 523   |

There are several methods for dealing with HLW. One of them is by transmuting the long-lived nuclear waste into shorter life by using Accelerator Driven Subcritical System (ADS) [8-11].

The Accelerator Driven Subcritical System is a system consisting of a subcritical nuclear reactor and a particle accelerator. Commonly, particle accelerators that used are cyclotron or linear accelerator (LINAC). The accelerated particles in an accelerator will hit target in subcritical reactor (k-eff <1). The reaction between these particles and the target is called a spallation reaction that produces a spallation product. Figure 1 shows the general schematic diagram of ADS [8].

![Figure 1. Schematic diagram of ADS [8]](image)

Through the transmutation process in ADS, we can reduce the radiotoxicity period of HLW by 1000 times and reduce the volume of residual fuel to 100 times. By carrying out this transmutation process it can convert long-lived radioactive elements into shorter-lived particles.
2. Methodology

In this preliminary study, we have evaluated the plutonium and minor actinides transmutation in ADS with 100 MWth and 200 MWth of output power. The fuel type that used in the study is U-ThO₂(NO₃)₃ which is added with plutonium (Pu) and minor actinides (MA). Initial fuel density of UO₂(NO₃)₃, PuMA(NO₃)₃, and HNO₃ can be calculated using the following equations which can be found in the reference [12]. Table 2 presents the required constants those used in equations (1) – (3) [12].

\[
\begin{align*}
a_i(T) &= a_i^0 (1 + a_i^1(T - 298.15) + a_i^{''}(T - 298.15)^2) \quad (1) \\
b_i(T) &= b_i^0 (1 + b_i^1(T - 298.15) + b_i^{''}(T - 298.15)^2) \quad (2) \\
\frac{\rho_m(T)}{\rho_o(T)} &= 1 + \sum_{i=1}^{3} a_i(T) C_i + \sum_{i=1}^{3} b_i(T) C_i^{3/2} \quad (3)
\end{align*}
\]

where:
- \( T \) : Temperature (K)
- \( \rho_m(T) \) : Density of the mixed aqueous solution (kg/m³)
- \( \rho_o(T) \) : Density of the pure solvent (water) at 298.15K (kg/m³)
- \( C_i \) : Concentration of \( i \) sub solvent (kmol/m³)
- \( a_i \) : Empirical constant
- \( b_i \) : Empirical constant

| \( i \) | Solute | \( a^0 \) | \( a' \) | \( a^{''} \) | \( b^0 \) | \( b' \) | \( b^{''} \) |
|---|---|---|---|---|---|---|---|
| 1 | U | 3.2271 \times 10^{-1} | -2.0720 \times 10^{-2} | 4.5448 \times 10^{-4} | -8.8561 \times 10^{-3} | -7.0788 \times 10^{-1} | 1.6824 \times 10^{-2} |
| 2 | Pu | 4.2405 \times 10^{-1} | -1.0963 \times 10^{-2} | -4.0997 \times 10^{-4} | -4.7204 \times 10^{-2} | 7.6038 \times 10^{-1} | -2.7268 \times 10^{-3} |
| 3 | HNO₃ | 3.8053 \times 10^{-2} | -2.0043 \times 10^{-2} | 4.9565 \times 10^{-5} | -2.6308 \times 10^{-3} | -1.0894 \times 10^{-1} | -6.6770 \times 10^{-5} |

The initial fuel density of UO₂(NO₃)₃, PuMA(NO₃)₃, and HNO₃ calculated using equations 1, 2 and 3 with the density of pure water at 298.15K is 997.0751 kg / m³ [12].

In this study we have evaluated two types of plutonium vector, those are: reactor grade plutonium (RGPu), and weapon grade plutonium (WGPu). The isotopic composition for RGPu was taken from the remaining composition of 3 GWth operated PWR with 33 GWd/ton burnup [13]. The mass of ratio of minor actinides to plutonium in the PWR spent fuel is 1:9 [13]. The isotopic composition of plutonium vectors and minor actinides (MA) vector can be found in Tables 3 - 5, respectively [13].

**Table 2. Constants for calculating the initial fuel density [12]**

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**Table 3. Reactor grade plutonium vector [13]**

| Pu-238 (%) | Pu-239 (%) | Pu-240 (%) | Pu-241 (%) | Pu-242 (%) |
|---|---|---|---|---|
| 1.58 | 57.76 | 26.57 | 8.76 | 5.33 |

**Table 4. Weapon grade plutonium vector [13]**

| Pu-238 (%) | Pu-239 (%) | Pu-240 (%) | Pu-241 (%) | Pu-242 (%) | Am-241 (%) |
|---|---|---|---|---|---|
| 0.01 | 93.8 | 5.8 | 0.13 | 0.02 | 0.22 |

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**Table 5. Minor actinides vector [13]**

| Np-237 (%) | Am-241 (%) | Am-243 (%) | Cm-242 (%) | Cm-243 (%) | Cm-244 (%) | Cm-245 (%) | Cm-246 (%) |
|---|---|---|---|---|---|---|---|
| 42.25 | 47.57 | 8.5 | 0.32 | 0.01 | 1.26 | 0.07 | 0.01 |

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The ADS core geometry in this study is a finite cylindrical shape with radius 147 cm and a height of 54,249 and a cladding thickness is 0.3 cm with stainless steels material.

In this study, we have utilized SRAC 2006 code developed by JAERI Japan to analyze reactor systems [14]. The nuclear data library is JENDL 4.0, which has a total nucleus of 406 isotopes [15].

3. Results and Discussion

Figure 2 shows the effective multiplication factor (k-eff) versus burn-up for RGPuMA spent fuel that charged in ADS with 100 MWth of output power. ADS can maintain its subcritical condition when the concentration of PuMA-Nitrate is above 62.6 kmol/m^3. When the concentration less than 62.6 kmol/m^3, the ADS becomes a critical assembly (nuclear reactor). This condition is also happened for RGPuMA spent fuel with 200 MWth of output power, as presented in Figure 3.

![Figure 2. K-eff of RGPuMA with thermal power 100 MWth](image1)

The effective multiplication factor as a function of burn-up for WGPuMA charged spent fuel in 100 MWth thermal power of ADS is given in Figure 4. If the concentration of PuMA-Nitrate is above 65.6 kmol/m^3, ADS can keep its subcritical condition. However, the ADS becomes a critical assembly if the concentration of PuMA-Nitrate is less than 65.6 kmol/m^3. The latter condition is also observed for WGPuMA spent fuel with 200 MWth of output power, as given in Figure 5. From these results, it can be concluded that we can transmute higher amount of WGPuMA spent fuel in ADS compared to RGPuMA spent fuel. This fact may due to the higher fraction of fissile plutonium (Pu-239 and Pu-241) in WGPuMA (93.93%) compared to that of RGPuMA (66.52%).

![Figure 3. K-eff of RGPuMA with thermal power 200 MWth](image2)
Figure 4. K-eff of WGPuMA with thermal power 100 MWth

Figure 5. K-eff of WGPuMA with thermal power 200 MWth

4. Conclusion

Preliminary study on transmutation of plutonium and minor actinides in ADS has been carried out. For RGPuMA, the ADS can maintained its subcritical condition when the concentration of PuMA-Nitrate is higher than 62.6 kmol/m³. For WGPuMA, the ADS can kept its sub-criticality for the amount of PuMA-Nitrate is larger than 65.6 kmol/m³. Higher concentration of the fissile isotopes in WGPu might be reason for larger amount of required PuMA-Nitrate in ADS.

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