

Evaluation on transmutation of minor actinides discharged from PWR spent fuel in the RSG-GAS research reactor

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Article history

Received 31 August 2018
 Revised 10 October 2018
 Accepted 29 November 2018
 Published Online 25 August 2019

Abstract

The evaluation of RSG-GAS research reactor for transmutation reactor was proposed to study its effectiveness to transmute minor actinides (MA), specifically Am-241, to support geologic storage/disposal. The Am-241 radionuclide was assumed to be discharged from 1000MWe PWR's spent fuel. The mass of Am-241 discharged from within a year operation of 1000MWe PWR was $1.65\text{E}+03$ gram, while the optimum Am-241 mass which can be transmuted in RSG-GAS - and still meet the safety requirements of reactivity - was $8.0\text{E}+03$ gram. This was equivalent to about cumulative Am-241 discharged from 5 units of 1000MWe PWR. In 10 cycles of RSG-GAS operation (about 2 years), the remaining of Am-241 is only about 100 grams. The ratio of Am-241 transmuted ($8.0\text{E}+03$ gram) and Am-241 produced in the RSG-GAS core ($1.98\text{E}-02$ gram) within 1-year operation shows the effectiveness of RSG-GAS as a transmutation reactor.

Keywords: Transmutation, minor actinide, RSG-GAS research reactor

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INTRODUCTION

Over the years, the utilization of nuclear power plant (NPP) to generate electricity has shown encouraging growth. Correspondingly, the production of radioactive waste and the inventory of spent fuel produced also increased. Since the spent fuel contains harmful long-lived radionuclides, proper management of these substances is necessary in order to protect the environment [1]. Previous study has reported that plutonium and the minor actinides (MA) are responsible for the radiotoxicity and heat generation of nuclear spent fuel. To minimize the long-term radiotoxicity risk, theoretical studies have been done to transmute MA such as Np-237, Am-241, Am-243, Cm-244, and Cm-245. Through this transmutation, these MA with high radioactivity and very long half-life will be transmuted short-lived fission products or stable species. This approach is widely practiced to support geologic storage/disposal usage.

The transmutation of radionuclides discharged from NPP spent fuel is made possible by prior technology of separation and partitioning and also fabrication of the target to be transmuted. These possibilities have been intensively studied [2-3]. Thermal reactors are widely studied in the case of transmuting MA. Apart from that, transmutation of MA in other systems has been widely studied such as boil water reactor, pressurized water reactor [4-12], fast reactor [13-15], accelerator-driven system [16], and fusion reactor [17,18]. Setiawan and Kuntjoro [19] proposed a preliminary approach to use high-flux RSG-GAS research reactor as a transmutation reactor for Am-241 of PWR's spent fuel in Asian region.

The objective of this study was to evaluate the transmutation of Am-241 radionuclide which was separated and partitioned from 1000MWe PWR spent fuel in the RSG-GAS research reactor. Among MA nuclides, Am-241 was selected for transmutation due to its long half-life. The evaluation includes the optimum mass capacity of Am-241 to be inserted and irradiated in the RSG-GAS core, while maintaining the negative reactivity to ensure the reactor's safety. The

transmutation Am-241 in the transmutation-fuel was compared with that of in the RSG-GAS fuel to observe the transmutation efficiency. At the end of this study, the transmutation of Am-241 greatly reduced the amount of nuclides which are harmful to humans and the environment in a short-term manner.

RSG-GAS as a transmutation reactor

RSG-GAS reactor is a plate type research reactor that has a low enrichment fuel of 19.75%. The reactor has been operated by Indonesian National Nuclear Energy Agency (BATAN) since 1987. To date, it has been operated normally and safely for more than 30 years, thanks to the aging management program which is conducted based on the requirements set by the regulatory body.

The reactor has 4 fuel elements (FE) and 8 control elements (CE). Each FE consists of 21 uranium silicide fuel plates ($\text{U}_3\text{Si}_2\text{-Al}$), whereas each CE consists of 15 uranium silicide fuel plates.

The configuration of the RSG-GAS core is shown in Figure 1. The reactor is operated at a power of 15MWt with a cycle length of 42 days. The Am-241 would be prepared as a cylindrical irradiation target with a height of 40 cm and a radius that varies according to the mass to be irradiated.

The core of RSG-GAS has 4 central irradiation positions (CIP) and other 4 irradiation positions (IP). The Am-241 target will be irradiated at the CIP located at the center of the reactor core, to receive a high neutron flux of $\sim 10^{14}$ n/cm².s so that it will rapidly decrease in number. The RSG-GAS core configuration was taken for reference in the calculations is the 97th RSG-GAS core (T-97). Referring to Figure 1, the first line in each fuel cell is the type (fuel or control element); the second line explains the burn-up (%) at EOC; and the third line describes the power peaking factor (PPF) at the EOC.

RSG-GAS is considered to be suitable with the requirement of transmutation reactor since it has a high neutron flux (in the order $\sim 10^{14}$ n/cm².s) and it has specific irradiation positions to be easily inserted with Am-241 target.

FE-01 0.00 1.018	FE-02 7.34 0.957	FE-03 27.03 0.915	FE-04 20.67 0.925	FE-05 28.5 0.834	FE-06 0.00 0.981		
FE-07 14.40 1.061	FE-08 46.83 0.906	IP	CE-01 44.55 0.977	FE-09 41.59 0.856	FE-10 33.11 0.875		
FE-11 6.91 1.125	FE-12 6.68 1.169	CE-02 16.28 1.189	FE-13 32.89 1.126	FE-14 45.63 0.949	CE-03 8.33 1.194	FE-15 39.98 0.916	FE-06 0.00 1.047
FE-17 12.91 1.063	CE-04 38.07 1.049	FE-18 21.35 1.193	CIP	CIP	FE-19 33.82 1.130	IP	FE-20 14.01 1.036
FE-21 26.69 1.000	IP	FE-22 43.65 1.037	CIP	CIP	FE-23 20.89 1.177	CE-05 31.20 1.090	FE-24 29.08 0.892
FE-25 7.10 1.029	FE-26 37.94 0.918	CE-06 0.00 1.224	F-27 38.61 1.000	FE-28 34.73 1.095	CE-07 24.05 1.110	FE-29 13.73 1.091	FE-30 0.00 1.083
FE-31 32.55 0.852	FE-32 44.82 0.813	CE-08 50.52 0.909	IP	FE-33 45.49 0.904	FE-34 19.92 0.988		
FE-35 0.00 0.946	FE-36 40.90 0.743	FE-37 20.81 0.952	FE-38 26.33 0.965	FE-39 14.46 0.941	FE-40 6.47 0.961		

Figure 1. The 97th Equilibrium core of RSG-GAS research reactor [20].

CALCULATION METHODOLOGY

In this current work, the selected PWR has 157 UO₂ fuels with a burn-up of 60GMWd/THM. The reactor has a cycle length of 18 months with heavy metal mass of 1.06E+05kg. This work included simulation calculations using several computer codes. Firstly, the ORIGEN2 code was used to determine the amount of Am-241 produced by the 1000MWe PWR reactor.

After the Am-241 mass was obtained, two assumptions were made: (i) The overall mass can be separated and fabricated into the irradiation target inside an aluminium cylinder clad. The cladding has a height of 40 cm with a radius corresponding to the amount of Am-241 target to be irradiated; (ii) The fuel was properly shielded to meet the occupational radiation protection requirements.

Next, the WIMS-D5 code was used to generate macroscopic cross-section for the target. Using the same code, calculations were carried out to find out the maximum target mass that can be irradiated inside the reactor core.

Furthermore, the reactor calculation to determine the amount of negative reactivity in the reactor due to insertion of Am-241 target was performed. The calculation was performed using the BATAN-2DIFF neutron diffusion code. In addition, the reactor calculations were also carried out to obtain the magnitude of neutron flux received by the target. The magnitude of neutron flux is useful in determining the duration needed by the target to be depleted after being irradiated in the reactor core. The calculation of the target run time in the reactor was done using ORIGEN2 code.

Calculations were also made to find out the transmutation-efficiency of RSG-GAS to transmute Am-241 target using the ORIGEN2 code. The transmutation-efficiency (TE) can be obtained by taking the ratio of Am-241 mass transmuted on the RSG-GAS core to the Am-241 mass produced from the reactor operation during the target burning time. This TE would be varied for different reactors. High TE value indicates the high effectiveness of a system as a transmutation reactor.

RESULTS AND DISCUSSION

From the calculation of 1000MWe PWR under equilibrium condition, the radionuclide mass for 1-year operation was obtained. Table 1 depicts that Am-241 had a mass of 1.65E+03 gram within a year of 1000MWe PWR operation.

Since it has a long half-life, this MA nuclide needs special handling, i.e.: sustainable geologic disposal and/or transmutation into stable nuclide. In this work, the transmutation was carried out by irradiating the nuclides in the reactor core to reduce the amount of nuclides rapidly. Transmutation Am-241 can be effectively done since this nuclide has a large thermal fission neutron absorption which is 1.395 atom/barn-cm. The proposed transmutation process was carried out at a 15MWt RSG-GAS research reactor that has CIP in the center of the core with the flux of about 10¹⁴ n/cm².s. The presence of high neutron flux caused Am-241 to be rapidly burnt and depleted.

Table 1. Mass of nuclides in PWR spent fuel.

No.	Nuclide	Total (kg)	Hal-life (year)
1	U-234	2.732E+01	2.47E+03
2	U-235	2.135E+03	7.10E+02
3	U-238	8.783E+04	4.51E+09
4	Np-237	2.597E+01	2.14E+06
5	Am-241	1.656E+00	4.58E+02
6	Am-242m	5.977E-02	1.52E+02
7	Am-243	2.775E+00	7.95E+03
8	Cm-253	1.196E-02	3.20E+01
9	Cm245	2.287E-02	9.30E+03
10	Cm-246	2.115E-05	5.50E+03

Using the WIMS-D5 code, the core calculation was carried out to determine the safety level of the reactor in terms of reactivity due to the presence of Am-241 target. The Am-241 target was irradiated inside aluminium tube with diameter of 1.20 cm and 40 cm height. Data on the macroscopic cross-section of WIMS-D5 calculation targets are presented in Table 2.

Table 2. Macroscopic cross-section of 1.7 gram Am-241 target (atom/b-cm).

Group	Removal	Absorption	Fission	Transp
1	1.18E+00	2.16E+00	0.00E+00	5.31E+00
2	6.97E-02	1.37E+00	0.00E+00	7.51 E+00
3	1.36E-01	3.68E-02	0.00E+00	6.32 E-01
4	9.64E-05	2.23E-02	0.00E+00	2.03 E+00

Group	Σ _{g→g}			
1	2.06 E+00	1.18 E+00	1.85E-03	6.91E-14
2	0.00 E+00	9.11 E+00	6.97E-02	7.45E-06
3	0.00 E+00	0.00 E+00	4.66E-01	1.36E-01
4	0.00 E+00	0.00 E+00	9.64E-05	2.31E+00

From Table 2, it can be seen that the macroscopic cross-section of the Am-241 target has large probability of forming interaction with neutrons in the thermal group. This is in accordance with the nature of the RSG-GAS which is a thermal reactor with a light water cooler.

Subsequent core calculations were performed for the RSG-GAS reactor core under conditions without and with Am-241 target in one CIP position. Reactor calculations were performed using the BATAN-2DIFF program. Further calculations were made for numerous variations of target mass Am-241. The calculation results are tabulated in Table 3.

Table 3 shows the Am-241 target of spent fuel 1000MWe PWR for 1-year operation of 1.70E+03 gram gives reactivity effect on RSG-GAS's core at -0.47%. This value does not have a harmful effect on the RSG-GAS reactor, since the reactor has a reactivity reserve for experiment or target irradiation of Δρ = ± 2%. For one position in CIP for a target of 6.00E+03 gram it is possible to irradiate (or transmute) since it only provides negative reactivity of ρ = -0.69%. Subsequent

calculations were performed by simulating to incorporate various Am-241 targets in two CIP positions to observe how much they would affect the RSG-GAS cores.

Table 3. k-eff and neutron flux of RSG-GAS for the initial condition.

No	Target (gram)	k-eff	Reactivity (%)	Flux (n/cm ²)
1	0	1.10729	0.00	1.232E+14
2	1000	1.10233	-0.45	1.207E+14
3	1700	1.10206	-0.47	1.205E+14
4	2000	1.10169	-0.51	1.204E+14
5	3000	1.10128	-0.54	1.202E+14
6	4000	1.10115	-0.55	1.201E+14
7	5000	1.10110	-0.56	1.201E+14
8	6000	1.09967	-0.69	1.194E+14

The results obtained were for 2 CIP positions which still meet the safety requirements of reactivity which is 8.00E+03 gram (2×4.00E+03 gram). This value is the maximum RSG-GAS can transmute the actinide minor. The amount of Am-241 that can be transmuted in the RSG-GAS core is relatively large. The difficulty faced is in target preparation, which requires radiation protection handling. Having this difficulty settled down, the advantageous will be obtained for Am-241 transmutation in the RSG-GAS. By transmutation for 2 years (10 cycles of operation), the remaining of Am-241 is only about 1.98E-02 gram. This is very beneficial, since the amount of Am-241 stored is very small in mass and volume.

The transmutation-efficiency, *TE*, is the ratio between the Am-241 transmuted per 1 year of operation (8.00E+03 gram) compared to the Am-241 produced in RSG-GAS core (1.98E-02 gram) as shown in Table 4. This *TE* value (4.04E+05) indicated that the RSG-GAS reactor was efficient in transmuting Am-241 of PWR waste products.

Table 4. Am-241 production in RSG-GAS core (in gram/year).

No.	Location	Radionuclide Am-241	
		(gram/cycle)	(gram/year)
1	CE-01	5.105E-08	3.063E-07
2	CE-02	9.741E-08	5.845E-07
3	CE-03	5.884E-06	3.530E-05
4	CE-04	1.893E-05	1.136E-04
5	CE-05	4.050E-05	2.430E-04
6	CE-06	8.887E-05	5.332E-04
7	CE-07	1.552E-04	9.312E-04
8	CE-08	2.461E-04	1.477E-03
1	FE-01	4.011E-08	1.203E-06
2	FE-02	8.552E-07	2.566E-05
3	FE-03	4.757E-06	1.427E-04
4	FE-04	1.645E-05	4.935E-04
5	FE-05	4.099E-05	1.230E-03
6	FE-06	8.084E-05	2.425E-03
7	FE-07	1.481E-04	4.443E-03
8	FE-08	2.579E-04	7.737E-03
TOTAL			1.983E-02

CONCLUSION

Calculation study to evaluate the transmutation of Am-241 discharged from PWR spent fuel using the available RSG-GAS research reactor was performed. This study revealed that the mass of Am-241 discharged from within a year operation of 1000MWe PWR was 1.65E+03 gram. The optimum Am-241 mass can be transmuted in RSG-GAS which still meet the safety requirements of reactivity was 8.00E+03 gram (or equal to about cumulative Am-241 discharged from 5 units of 1000MWe PWR). In 2 years (10 cycles of operation)

transmutation, the remaining of Am-241 was only about 100 grams. The mass of Am-241 transmuted (8.00E+03 gram) produced in the RSG-GAS core (1.98E-02 gram) within 1 year demonstrated the effectiveness of RSG-GAS as a transmutation reactor.

ACKNOWLEDGEMENT

This research was financially supported by the Head of PTKRN, BATAN.

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