Evaluation of Potential Scenarios for Weapon-Grade Plutonium from MAGNOX Type Reactor

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1. Introduction

For the global nuclear nonproliferation, it is important to figure out the capability of surrounding countries for producing nuclear bombs. One of the key factors is the production of weapon-grade (WG) plutonium for nuclear weapons. In general, the plutonium having higher Pu²³⁹ content than 93 wt% is classified as the WG plutonium. Various reactors including graphite and heavy-water moderated reactors can be used for high quality plutonium production. Most estimation of the WG plutonium production were performed using a very simple formula or point depletion models. Recently, the authors analyzed the plutonium production for a 5MWe MAGNOX type reactor using MCNP quarter core model when natural uranium was loaded in graphite-moderated gas-cooled reactor [1].

However, surrounding countries are likely to prefer to produce more amount of WG plutonium through their reactors and so there are possibilities to change the core loading pattern for more effective WG plutonium production. In this study, we considered the several potential scenarios by changing the fuel rod enrichment in a central region of 5MWe MAGNOX type reactor core [1]. Then, the plutonium production and Pu²³⁹ content were analyzed versus the fuel enrichment and central region size. In particular, we suggest which scenario produces the largest amount of WG plutonium assuming full core reprocessing.

2. Modeling and methods

2.1 Graphite-moderated gas-cooled reactor model

The main design of the graphite-moderated gas-cooled reactor was organized in the previous study based on the Kang's work [2]. However, slight changes are made on the diameter of control rod channel such that it is changed from 13 cm to 6.5 cm and a little beryllium is added to the cladding composition to consider more realistic data. But these changes do not affect the core reactivity and other results. The reactor is classified as the Magnox type because it uses the Magnox cladding (99% Mg) and uses CO2 gas as the coolant. The core consists of 812-877 fuel channels and a single fuel channel consists of 10 fuel rods axially. We adopted 801 fuel channels such that the initial uranium loading would be 50 tons. The effective core height is 592 cm and each fuel rod is 60 cm long. Each fuel rod having 0.05 cm thick cladding contains 6.24 kg natural uranium. There are 44 control rod channels to control reactivity. 300 tons of graphite compose moderator and reflector. In particular, it is noted that this reactor has a very low specific power of ~0.5 MWt/tHM due to its low thermal power (i.e., 25MWt) and large uranium loading. This specific power is much lower by a factor of 0.013 than those of the typical PWRs [1].

2.2 Methods

The main design of graphite-moderated gas-cooled reactor mentioned in section 2.1 was modeled on quarter core symmetry using Monte Carlo code MCNP6 [3]. For detailed depletion analysis, we treated each of 10 axial fuel zones for each fuel channel as the depletion zone in the MCNP6 calculations, which led to total 2140 depletion zones. In addition, we modelled the empty and control channels as the same zone filled with CO_2 coolant for simplicity and all the external regions outside the core was modelled as a single graphite reflector region [1].

In this study, we considered several potential scenarios varied from modeling in section 2.1. The fuel channels with different uranium enrichment were loaded in the core central region for each scenario. The natural uranium (0.71 wt% U²³⁵) was used in the fuel channels of the remaining outer region. The following six different uranium enrichments in the central region are considered: 1) depleted uranium (0.2 wt%), 2) 1 wt%, 3) 2 wt%, 4) 3 wt%, 5) 4 wt%, and 6) 5 wt%. As mentioned above, each channel has 10 fuel rods loaded axially, and the uranium enrichment within each channel was assumed to be the same. In addition, we considered three different areas of the central region to show the effect of the central region area having different uranium enrichment from natural uranium on the WG production. Fig. 1 shows the quarter core models for three different central region areas. For these cases of the central region area, the central region is comprised of 41 channels, 133 channels, and 485 channels, respectively, on the full core. Therefore, the total number of the scenarios or cases is 19 (6 fuel enrichment cases for each of 3 central channel region cases plus one reference case loaded with a single natural uranium fuel channel). For these 19 cases, we analyzed the trend of Pu²³⁹ contents and plutonium productions.

2.3 Development of Analysis Tool for Magnox MCNP Model

The MCNP input preparation and analysis of the MCNP output for Magnox core require significant effort and time. Also, there are some possibilities that some errors or mistakes can be included in the modeling and analysis. Therefore, we developed a program called MATMM (Modeling and Analysis Tool for Magnox using MCNP) to prepare MCNP input file easily and

concisely for each scenario and to provide the target physical quantities after automatically processing the corresponding MCNP output file. The program was developed based on C# and supports Graphical User Interface (GUI) to make it easier for users to set up and to analyze scenarios.

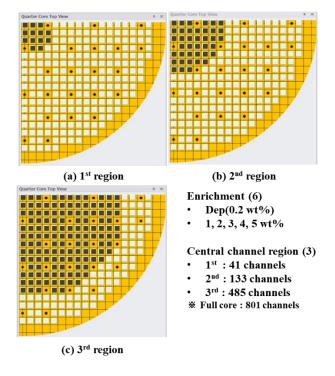


Fig. 1. Quarter core configurations for three different central region cases

For modeling using MATMM, users can easily set uranium enrichments and temperatures for target locations and edit Monte Carlo options including the important parameters such as reactor thermal power and operating period. Although only 19 scenarios are considered in this study, users can easily create MCNP6 inputs for various cases through this program. Fig.2 shows an example of creating a sample scenario. The user can name the channels and set the uranium enrichments for axially 10 fuel rods on the channels. After all the channel types are set or defined, the fuel channels can be loaded in some desired positions by clicking or dragging the desired position in the right 1/4 core plot. In addition, Monte Carlo options and reactor history input parameters can be easily set with MATMM. The Monte Carlo options include the number of histories per cycle, the number of total cycles, and the number of inactive cycles while the reactor history input parameters include the reactor thermal power and the depletion time steps.

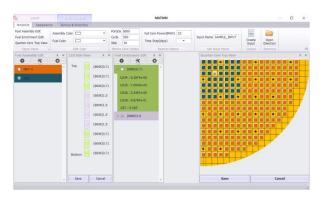


Fig. 2. Overview of creating a sample scenario with MATMM

After creating the MCNP6 input with MATMM, Monte Carlo depletion calculations provide burnup (GWd/MTU), mass (gram) and radioactivity (Ci) for 2,140 depletion zones in the output file. Extracting and analyzing the data for 19 scenarios also take a lot of effort and time. So, we added a module to MATMM that helps analysis of the data by importing MCNP6 output files. After an MCNP6 output file is imported, the user can plot the graphs by selecting the target parameters and the plotting options such as depletion time step. Fig. 3 illustrates an example for plotting a graph, in which the x-axis is set to days and y-axis to mass inventory. The nuclides to be plotted were set using their numeric identifiers (e.g., 94239 for Pu²³⁹ and 94240 for Pu²⁴⁰) and the desired depletion zones to be analyzed can be also set in the program.

When the MCNP6 output file is imported from MATMM, it is automatically saved as a JSON file. JavaScript Object Notation (JSON) is a lightweight, textbased, language-independent data interchange format based on JavaScript object syntax. Even though it closely resembles JavaScript object literal syntax, it can be used independently of JavaScript, and many programming environments feature the ability to read (parse) and generate JSON [4]. Because it takes a long time to extract data from the MCNP6 output file, the extracted data is saved as a JSON format file. The next time users analyze the same data, users can import the JSON file to extract the data quickly. Plotted graphs can be used for data analysis by copying or saving data.



Fig. 3. Overview of MATMM data analysis module

3. Results

First, we selected the feasible scenarios based on the evolution of the effective multiplication factor (keff). The scenarios with an initial effective multiplication coefficient (keff) of 1.0 to 1.2 were selected because too large excess reactivity cannot be compensated using the existing control rods. Fig.4 shows the evolutions for keff as depletion time for all the considered scenarios. In Fig.4. the cases for different central region area are designated using different symbols, while the ones for different the uranium enrichments are designated using different colors. Specifically, the smallest central channel region case (i.e., 1st case) is designated by circles, while 2nd and 3rd cases are designated by triangles and diamonds, respectively. The squares designate the reference case loaded with only natural uranium, represented as "Normal" and the reference case data is come from our previous studies [1]. As a result of selecting operable scenarios, only six cases were selected for further analysis: 1) depleted U, 1.0 wt% and 2.0 wt% uranium enrichment cases for the smallest central region case, 2) 1.0 wt% uranium enrichment case for the second central region area case, and 3) 1.0 wt% uranium enrichment case for the third central region area case.

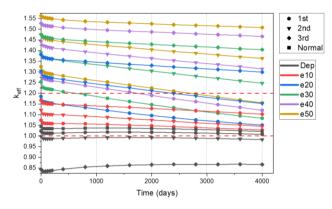


Fig. 4. Comparison of the evolutions of k_{eff} as depletion time

Next, the evolution characteristic of Pu²³⁹ content over depletion time was analyzed for all scenarios. The Pu²³⁹ content was calculated for the entire core and it is defined the ratio of Pu^{239} to the total plutonium mass (Pu^{236} ~ Pu²⁴²). Fig.5 shows the Pu²³⁹ content over depletion time for the different central region area cases. As shown in Fig.5(a), loading depleted uranium in the 1st region gives highest Pu²³⁹ content over depletion time. As the fuel enrichment increases, the Pu²³⁹ content gradually decreases and reaches the lowest point between 4% and 5%. In the 2^{nd} central region case, as shown in Fig. 5(b), 5% enriched uranium loading in the central region gave the highest Pu²³⁹ content. As the fuel enrichment decreases in the central region, the Pu²³⁹ content decreases and reaches the lowest point between 1% and 2%. In the case of depleted uranium loading in the central region, the Pu²³⁹ content increased slightly compared to

the case of 1% enriched uranium loading case. In the last central region case, as shown in Fig.5(c), the highest Pu^{239} content was evaluated for 5% enriched uranium loading. The lower the fuel enrichment, the lower the Pu^{239} content, depending on the depletion period.

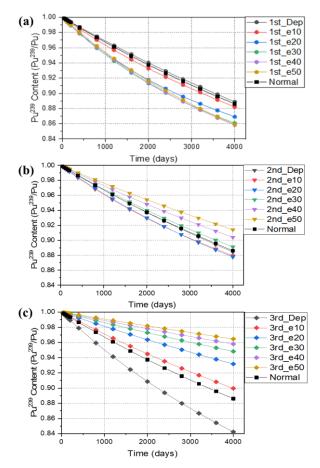


Fig. 5. Comparison of evolution of Pu²³⁹ content as depletion time for 19 scenarios.
(a): 1st region, (b): 2nd region, (c): 3rd region

Next, the trend of plutonium production in the entire core over depletion time was analyzed. The results are shown in Fig.6. In all three graphs, loading depleted uranium in the central region was estimated to give the highest plutonium production. Comparing the plutonium mass of all scenarios, the scenario loaded with depleted uranium in the largest central region gave the highest plutonium production. In addition, the higher the fuel enrichment, the lower the plutonium production depending on the depletion period. Furthermore, when the fuel enrichment is higher than natural uranium, the plutonium production becomes smaller as the amount of fuel loaded increases. On the other hand, if the amount of fuel with lower enrichment than natural uranium increases, the plutonium production increases.

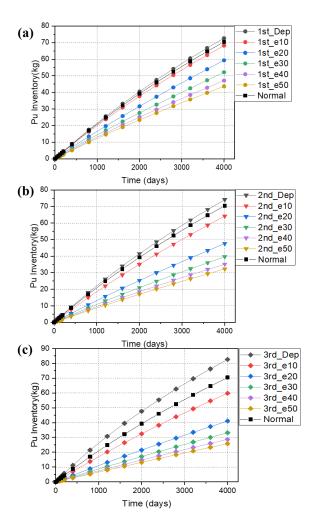


Fig. 6. Comparison of evolution of Pu inventory as depletion time for 19 scenarios.
(a): 1st region, (b): 2nd region, (c): 3rd region

Finally, we tried to find the scenario giving the largest amount of WG plutonium production. Fig. 7 shows simultaneously Pu²³⁹ content and plutonium inventory over depletion time by scenario, assuming reprocessing of the entire core. Since the Pu²³⁹ content decreases over depletion time in Fig. 5 and the plutonium production increases over depletion time in Fig.6, the graph for each scenario in Fig.7 progresses upward over depletion time. The higher the plutonium inventory at the same Pu²³⁹ content, the better the scenario can be determined. In other words, the larger the slope, the better the scenario. According to the graph, the scenario of "3rd_e50" is the best scenario giving the largest slope. However, for each scenario in Fig.7, the dot corresponds to 1600 days (about burnup of 800 MWd/MTU) of operation, and the "3rd_e50" scenario is high in Pu²³⁹ content but low in plutonium production than other scenarios in comparison to the same period. Therefore, the "1st Dep" and "2nd Dep" scenarios are the scenarios giving large amount of WG Plutonium production. That is to say, they have higher plutonium production and higher Pu²³⁹

content based on the same operating period than the other scenarios. With considering only the operable scenario from viewpoint of excess reactivity, "1st_Dep" is evaluated as the scenario giving the largest amount of WG plutonium production. It is noted that this scenario shows higher Pu²³⁹ content and plutonium production than the reference ("Normal") scenario loaded with only natural uranium. In this scenario, 93% Pu²³⁹ content is reached after depletion at about 2265 days, with a producible plutonium inventory of 43.73 kg. However, since this study evaluated the Pu²³⁹ content over the entire core, it is expected that some fuel channels can contain Pu²³⁹ contents lower than 93%. Therefore, the actual amount of WG plutonium inventory obtained from reprocessing is expected to be lower than the analysis above.

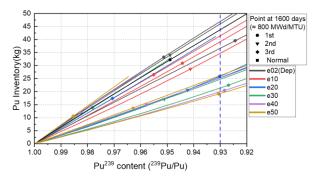


Fig. 7. Comparison of plutonium inventory as Pu²³⁹ content for 19 scenarios.

3. Conclusions

In this study, plutonium production and Pu²³⁹ content analysis were performed with MCNP core depletion analysis for several scenarios for a 5MWe MAGNOX reactor which is graphite-moderated gas-cooled. The scenarios were set by considering the fuel channels with different fuel enrichments at the central region of the reactor. Totally, 18 scenarios and one reference scenario loaded only natural uranium fuel were set up, dividing the central region area into three cases and the uranium enrichment into six cases. Because of consideration of many scenarios and depletion zones, we developed a program (MATMM) to automatically prepare MCNP input files and analyze MCNP output files. All results were analyzed by applying this program. From viewpoint of the excess reactivity, six scenarios were selected as the operational scenarios. Next, trend of the Pu239 content and plutonium produced in the entire reactor were analyzed for all the scenarios. For all the scenarios, the Pu²³⁹ content tended to decrease over time. The evolution characteristics of Pu²³⁹ content varies strongly depending on the central loading channel region and uranium enrichment in the central region and the highest Pu²³⁹ content over depletion time was shown for the case that highest enriched uranium fuel is loaded in the largest central region, but this case has very high excess reactivity. In the case of plutonium production, it has

been found that plutonium production increases as the amount of fuel with a lower enrichment than natural uranium increases. Thus, the largest amount of plutonium production was shown in the case that the depleted uranium is loaded in the largest central region, but this case has no excess reactivity. With the consideration of excess reactivity, the case loaded with depleted uranium in the small central region was shown to produce largest amount of WG plutonium.

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