Estimation of Plutonium Production in a Graphite-moderated Reactor using Graphite Isotope Ratio Method and MCS

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1. Introduction

These days, the international community, and especially the South Korean government, is making efforts to denuclearize the North Korea. If North Korea accepts a complete denuclearization, it would be essential to estimate the amount of plutonium produced by them. This estimation would help to determine the number of produced plutonium nuclear weapons, by that verifying the status of denuclearization. It is known that the North Korea is producing weapon-grade plutonium using a Magnox-type reactor in Yongbyon [1, 2]. The amount of plutonium produced in a Magnox reactor can be estimated using a correlation between the ratio of impurity indicator isotopes and the generated plutonium.

This paper presents an estimation of ^{239}Pu production in a graphite-moderated nuclear reactor using a Graphite Isotope Ratio Method (GIRM) [1, 3] paired with our code MCS [4]. MCS is a continuous-energy Monte Carlo code developed at the Computational Reactor Physics and Experiment laboratory (CORE) of Ulsan National Institute of Science and Technology. In this study, the ratio of ^{10}B/^{11}B isotopes was used as the impurity indicator, while MCS was employed to perform the Magnox reactor depletion calculation [5].

2. Methods

In this section, the geometry of Magnox reactor is provided, and the detailed process of GIRM is explained.

The concept of GIRM is that the impurity isotope ratio change caused by transmutation is proportional to the cumulative plutonium production. However, since the impurity isotope ratio is different for each region in the 3D whole core and the ratio data for each region is limited, a least-squares regression is applied in order to compensate the lacking data.

2.1 Magnox Reactor

Magnox reactor is using natural uranium as fuel, graphite as moderator and carbon dioxide (CO_2) gas as the heat exchange coolant. It was designed to produce both electric power and weapon-grade ^{239}Pu. Fig. 1 and Table I present the radial layout and design parameters of Magnox reactor, respectively.

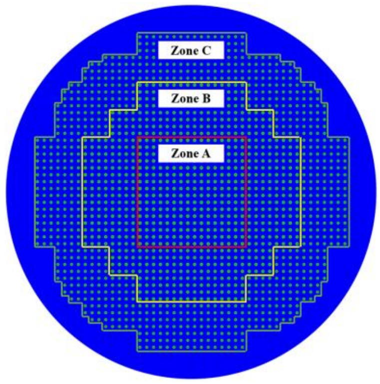

Fig. 1. Radial layout of Magnox reactor.

Table I: Design parameters of Magnox reactor

Parameter		Value	Unit
Power		182	MW_{th}
Active height		640	cm
Active diameter		945	cm
Fuel pin radius		1.4610	cm
Cladding radius		2.0400	cm
Coolant radius	Zone A	5.2080	cm
	Zone B	5.0165	
	Zone C	4.5847	
Fuel temperature		800	K
Moderator temperature		650	K

2.2 MCS depletion simulation

The depletion simulation of Magnox reactor was performed using MCS. The effective multiplication factor change during depletion is presented in Fig. 2.

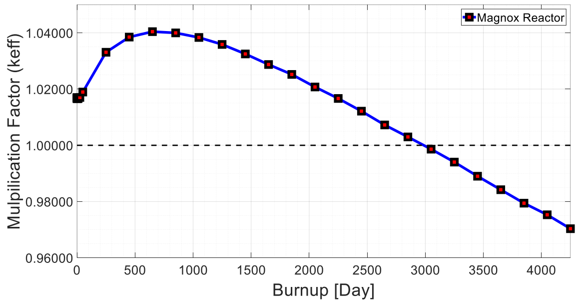

Fig. 2. Effective multiplication factor.

Standard deviation for the multiplication factor in this calculation was found around 20 pcm. Cumulative ^{239}Pu production for each depletion step obtained using MCS is shown in Fig. 3. This data is used further as the reference data.

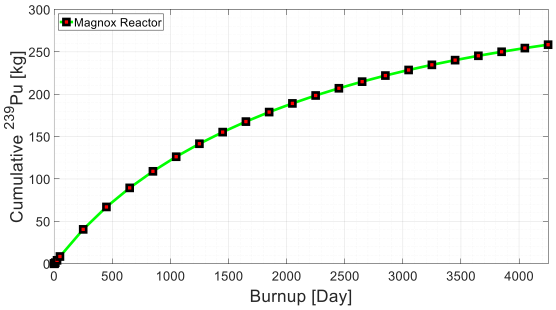

Fig. 3. Cumulative ^{239}Pu production.

2.3 Graphite Isotope Ratio Method (GIRM)

The detailed process of GIRM applied in this study is the following. First, ^{239}Pu mass density for the ^{10}B/^{11}B ratio is calculated for each depletion step using MCS 2D fuel pin simulation. Then, the ^{239}Pu mass density is estimated using a corresponding 2D fuel pin ^{10}B/^{11}B ratio for each sampling region of a 3D whole core simulation. Alternatively, ^{10}B/^{11}B ratio calculation can be replaced by measurement data. A 3D space-dependent equation of ^{239}Pu mass density for the whole core is derived through a least-squares regression using a ^{239}Pu mass density for each sampling region. Finally, the total estimated ^{239}Pu production is calculated by integrating the equation over the whole core 3D space. The accuracy of GIRM is evaluated by comparing the total ^{239}Pu production calculated using MCS and GIRM. Fig. 4 presents the flowchart of GIRM process. Fig. 5 shows the ^{239}Pu mass density for the ^{10}B/^{11}B ratio in a 2D fuel pin.

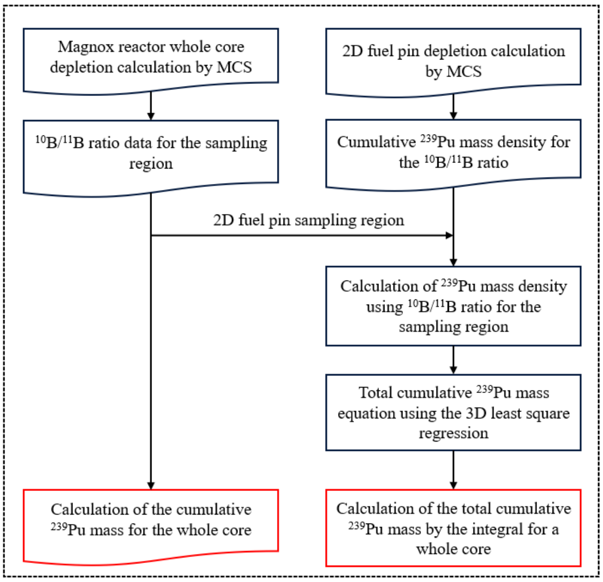

Fig. 4. Flowchart of GIRM process.

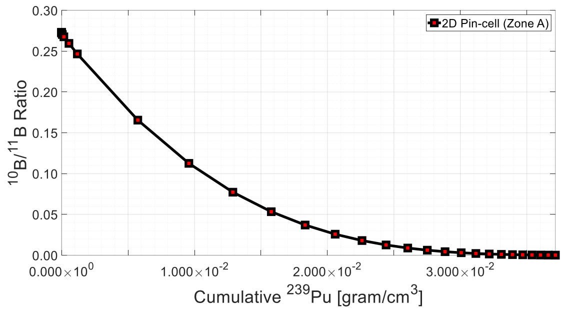

Fig. 5. ^{239}Pu mass density for the ^{10}B/^{11}B ratio in a 2D fuel pin.

Finally, the radial and axial sampling regions of the Magnox core are given in Fig. 6. Since the configuration of fuel pins in a whole core has a quarter-core symmetry, the sampling regions were chosen within the quarter core. The number of radial and axial sampling regions is set as 28 and 5, respectively. Therefore, a total of 140 sampling region data were used.

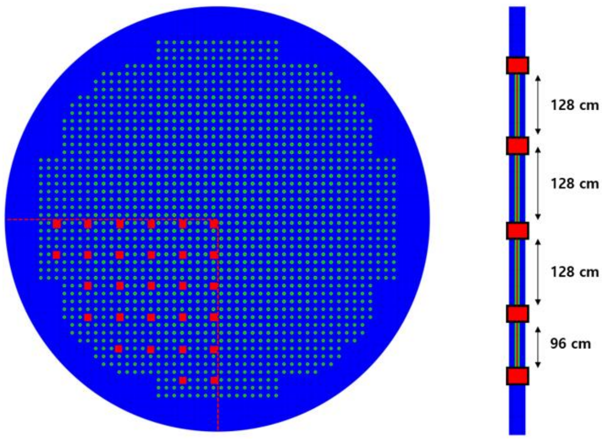

Fig. 6. Axial and radial sampling regions.

3. Results

In this section, the results of the ^{239}Pu production obtained by both MCS simulation and GIRM estimation are provided and compared.

3.1 Total Cumulative ^{239}Pu production

As explained in section 2.3, a 3D space-dependent least-squares regression function based on the triangular basis was used to approximate the ^{239}Pu mass density for the whole core as shown in Eq. (1).

$$f(x,y,z) = \sum_{k=0}^{z^{order}} \sum_{\substack{i=0 \\ j=0}}^{i+j\le xy^{order}} a_{i,j,k} x^i y^j z^k \qquad \text{Eq. (1)}$$

where $f(x,y,z)$ is the ^{239}Pu mass density for the *(x,y,z)* location in a whole core, z^{order} and xy^{order} are the regression orders for z and xy, respectively. In this study, several orders of *z* and *xy* were tested in order to find the optimized results as shown in Fig. 7. Each case result was compared to the reference data calculated by MCS. Through this process, the 3rd order of *z* and *xy* shows the smallest root mean square (RMS) error.

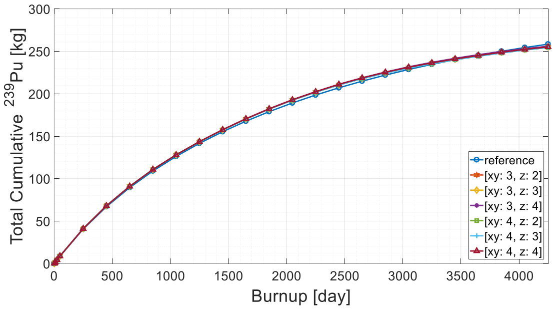

Fig. 7. Finding the optimal order of *z* and *xy* for least-squares regression.

Table II and Fig. 8 present a comparison of the total cumulative ^{239}Pu production calculated using MCS and GIRM.

Table II: Comparison of total cumulative ^{239}Pu production calculated by MCS and GIRM

Burnup [day]	MCS	Estimated	Relative Error [%]
	Total Cumulative ^{239}Pu [kg]		
0	0.00	0.00	0.000
50	8.51	8.77	3.036
250	40.53	41.07	1.336
450	66.99	68.09	1.653
650	89.49	90.98	1.657
850	109.03	110.82	1.643
1050	126.25	128.10	1.461
1250	141.58	143.54	1.384
1450	155.31	157.55	1.441
1650	167.70	170.40	1.609
1850	178.94	182.07	1.749
2050	189.17	192.61	1.816
2250	198.53	201.99	1.748
2450	207.03	210.62	1.734
2650	214.86	218.07	1.494
2850	222.04	224.81	1.249
3050	228.63	230.86	0.975
3250	234.68	236.16	0.629
3435	240.24	240.94	0.291
3650	245.38	245.20	-0.075
3850	250.10	249.08	-0.410
4050	254.46	252.40	-0.812
4250	258.46	255.23	-1.250

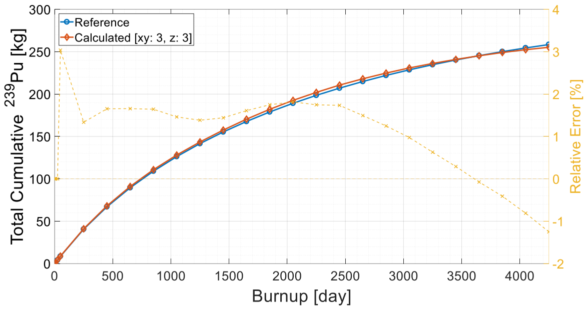

Fig. 8. Comparison of total cumulative ^{239}Pu production calculated by MCS and GIRM.

3.2 Axial and Pin-wise Cumulative ^{239}Pu production

To evaluate the space dependence effect of the produced result, the ^{239}Pu production for various axial regions and fuel pins for the depletion step of 3250 days were compared. A comparison of the axial cumulative ^{239}Pu production calculated with MCS and GIRM is shown in Table III.

Table III: Comparison of axial cumulative ^{239}Pu production calculated by MCS and GIRM on depletion step of 3250 day

Height [cm]		MCS	Estimated	Relative Error [%]
Bottom	Top	Total Cumulative ^{239}Pu [kg]		
100	132	9.208	10.047	9.114
132	164	10.421	10.586	1.579
164	196	11.174	11.069	-0.946
196	228	11.682	11.496	-1.594
228	260	12.045	11.865	-1.487
260	292	12.301	12.178	-1.005
292	324	12.477	12.431	-0.370
324	356	12.607	12.625	0.142
356	388	12.681	12.759	0.612
388	420	12.719	12.832	0.884
420	452	12.725	12.843	0.921
452	484	12.689	12.791	0.808
484	516	12.606	12.676	0.560
516	548	12.485	12.497	0.097
548	580	12.309	12.253	-0.459
580	612	12.056	11.943	-0.944
612	644	11.691	11.566	-1.072
644	676	11.176	11.122	-0.484
676	708	10.423	10.609	1.784
708	740	9.209	10.028	8.891

As for the pin-wise cumulative ^{239}Pu production, several fuel pin locations in different regions of the core were picked. The chosen locations are shown in Fig. 8. The comparison of cumulative ^{239}Pu production calculated using both MCS and GIRM for each chosen fuel pin is presented in Table IV.

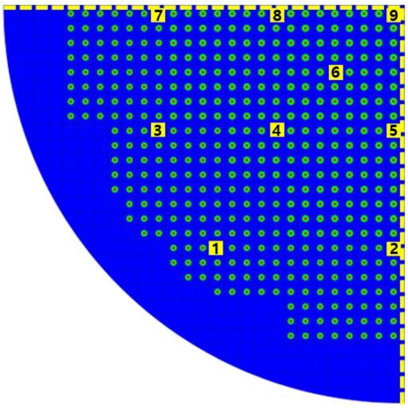

Fig. 9. Fuel Pin Index for Pin-Wise Comparison of ^{239}Pu production.

Table IV: Comparison of cumulative ^{239}Pu production calculated with MCS and GIRM for each chosen fuel pin after 3250 days of operation.

Fuel Pin Index	MCS	Estimated	Relative Error [%]
	Total Cumulative ^{239}Pu [kg]		
1	0.121	0.124	2.704
2	0.145	0.148	2.205
3	0.135	0.137	1.570
4	0.155	0.154	-0.468
5	0.157	0.155	-1.093
6	0.158	0.153	-3.385
7	0.144	0.146	1.350
8	0.158	0.154	-2.112
9	0.161	0.156	-2.940

4. Conclusions

In this study, the amount of ^{239}Pu production in a graphite-moderated Magnox reactor was estimated using MCS and GIRM and the results produced by those two methods of calculation were compared.

As presented in Table II, the total cumulative ^{239}Pu production calculated by MCS and GIRM show a difference of 1.157% RMS on average, with the maximum and the minimum RMS of 3.306% and -1.250%, respectively. In addition, the space dependence of the ^{239}Pu estimation was evaluated by comparing the cumulative ^{239}Pu production for various axial regions of the core as well as individual fuel pins. The result was found within acceptable range.

The future work will be applying other impurity indicator isotope ratios such as ^{6}Li/^{7}Li, ^{48}Ti/^{49}Ti and ^{235}U/^{238}U. In addition, more sensitivity tests for the number and position of sampling regions will be performed because practical location of sampling region in the actual reactor is unclear.

Acknowledgments

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