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Verification of Graphite Isotope Ratio Method Combined With Polynomial Regression for the Estimation of Cumulative Plutonium Production in a Graphite-Moderated Reactor

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Graphite Isotope Ratio Method (GIRM) can be used to estimate plutonium production in a graphite-moderated reactor. This study presents verification results for the GIRM combined with a 3-D polynomial regression function to estimate cumulative plutonium production in a graphite-moderated reactor. Using the 3-D Monte-Carlo method, verification was done by comparing the cumulative plutonium production with the GIRM. The GIRM can estimate plutonium production for specific sampling points using a function that is based on an isotope ratio of impurity elements. In this study, the $^{10}B/^{11}B$ isotope ratio was chosen and calculated for sampling points. Then, 3-D polynomial regression was used to derive a function that represents a whole core cumulative plutonium production map. To verify the accuracy of the GIRM with polynomial regression, the reference value of plutonium production was calculated using a Monte-Carlo code, MCS, up to 4250 days of depletion. Moreover, the amount of plutonium produced in certain axial layers and fuel pins at 1250, 2250, and 3250 days of depletion was obtained and used for additional verification. As a result, the difference in the total cumulative plutonium production based on the MCS and GIRM results was found below 3.1% with regard to the root mean square (RMS) error.

Keywords: Graphite Isotope Ratio Method, GIRM, MCS, Plutonium production, Non-proliferation

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1. Introduction

The cumulative plutonium production is proportional to the neutron fluence and so does the change of the impurity elements in the graphite moderator. So, if we know the change of the impurity elements in the graphite moderator, we can estimate the cumulative plutonium production. In many cases, however, we don't know the change of impurity elements because the initial amount of impurities in the graphite varies from graphite to graphite and is usually unknown.

To overcome this difficulty, GIRM was developed at Pacific Northwest National Lab (PNNL) in 1990's [1, 2]. Although the initial amount of impurity elements is unknown, the isotopic ratios of the impurity elements such as $^{10}B/^{11}B$ and $^{36}Cl/^{35}Cl$ are known initially and their change does not depend on the amount of impurity elements but depends only on the neutron fluence. Once the cumulative plutonium production is tabulated as a function of the isotopic ratio of an impurity element using a simple two-dimensional (2-D) unit fuel pin cell model, the cumulative plutonium production can be estimated by measuring the isotopic ratio of the impurity elements in the graphite moderator of the reactor of interest under the assumption that the correlation between the cumulative plutonium production and the isotopic ratio of the impurity elements from the unit fuel pin cell model is applicable everywhere in the core.

In our previous work, a suitability study was done for many candidate indicator isotopes of impurity elements in the graphite moderator, and it was found that $^{10}B/^{11}B$, $^{36}Cl/^{35}Cl$, $^{48}Ti/^{49}Ti$ and $^{235}U/^{238}U$ have a consistent correlation with the cumulative plutonium production, regardless of the initial impurity concentration of the graphite. On the other hand, the correlation between $^{6}Li/^{7}Li$ and plutonium production depends on the initial concentration of the boron impurities in the graphite because ^{7}Li can be produced both by the neutron capture reaction of ^{6}Li and by the (n, α) reaction of ^{10}B [3].

When GIRM is applied to estimate the cumulative plutonium production of a graphite-moderated reactor, isotopic ratio measurements of impurity elements are not performed for all fuel channels for practical reasons but for some fuel channels. In this case, the cumulative plutonium production of the whole core should be estimated from the measured isotopic ratios. A 3-D cumulative plutonium production map can be produced by a 3-D regression based on the measured isotopic ratios [4].

In this work, the accuracy of a 3-D polynomial regression technique for estimating cumulative plutonium production in a graphite-moderated reactor is assessed. As the reference reactor, a Magnox-type reactor, British Calder Hall reactor [5] was selected. The 3-D depletion calculation for the reference reactor was performed using the MCS code [6], a continuous-energy neutron transport Monte-Carlo code, developed at the COmputational Reactor physics and Experiment laboratory (CORE) of Ulsan National Institute of Science and Technology (UNIST). From the 3-D depletion calculation of the reference reactor, the total cumulative plutonium production of the whole core was calculated for every burnup steps and the result are taken as the reference one. On the other hand, the cumulative plutonium production of the whole core was estimated using the 3-D polynomial regression technique and it was compared with the reference result. In the 3-D regression technique, the cumulative plutonium production was estimated using the $^{10}B/^{11}B$ ratio at some points from the reference 3-D depletion calculation and the tabulated cumulative plutonium production as a function of $^{10}B/^{11}B$ ratio using the unit fuel pin cell model of the reference reactor.

2. Method

In this section, the specifications of Calder Hall reactor used in this study and the depletion calculation results for the reactor using MCS are provided. Also, the process of GIRM combined with polynomial regression is explained.

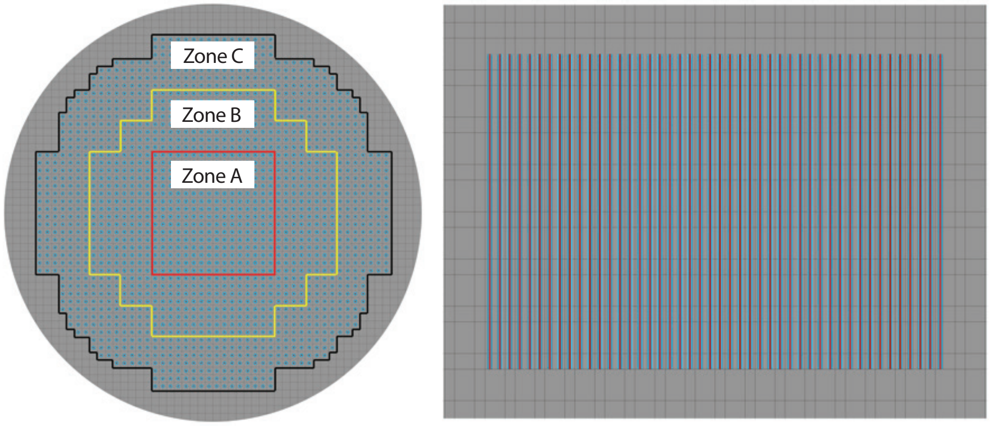


Fig. 1. Radial and axial layout of Calder Hall reactor.

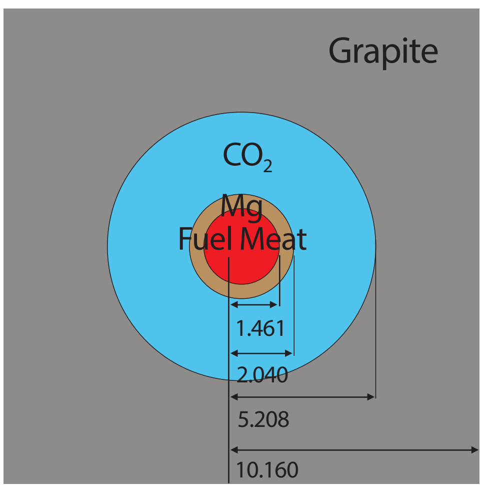


Fig. 2. Geometry of fuel pin for Zone A.

Table 1. Design parameters of Calder Hall 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 hole radius	Zone A	5.2080	cm
	Zone B	5.0165	
	Zone C	4.5847	
Fuel temperature		800	K
Moderator temperature		650	K
Fuel density		17.98	$g{\cdot}cc^{-1}$
Cladding density		1.65	$g{\cdot}cc^{-1}$
Number of fuel pin		1,696	-

2.1 Calder Hall Reactor

The detailed specifications of the reactor used to produce plutonium in Yongbyon nuclear scientific research center of North Korea are unknown. However, it is known to be a smaller version of British Calder Hall reactor [7]. Therefore, the Calder Hall reactor whose specifications are well known was taken as the reference reactor in this study. The Calder Hall reactor was used to produce plutonium as well as to produce electricity as the world's first commercial reactor. The fuel, the moderator, and the coolant of the reactor are natural uranium metal, graphite, and carbon dioxide (CO_2) gas, respectively. The core has three zones, Zone A, Zone B, and Zone C and the radius of the coolant hole is different for each zone. The radial and axial layout of Calder Hall reactor are presented in Fig. 1, the geometry of fuel pin for Zone A is shown in Fig. 2. Also, Table 1 presents the design parameters.

The depletion simulation of the Calder Hall reactor was

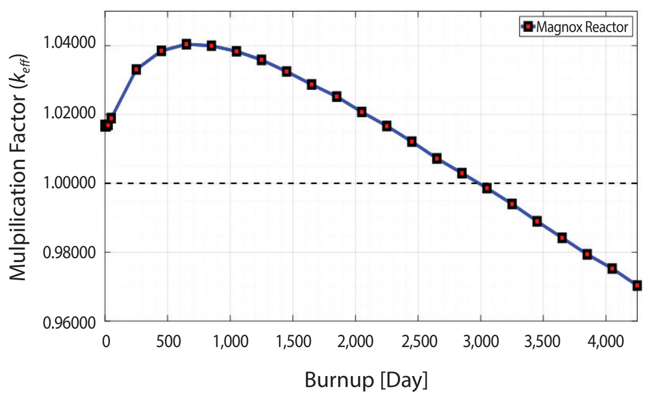


Fig. 3. Effective multiplication factors of whole core.

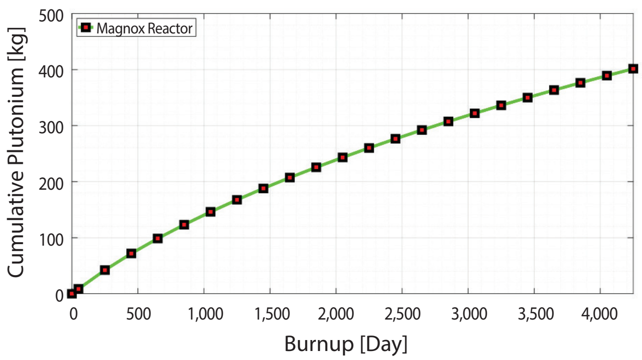


Fig. 4. Cumulative plutonium production for the Calder Hall reactor.

performed using MCS with a nuclear cross-section library based on ENDF/B-VII.1 and HELIOS kappa library. The effective multiplication factors (k_{eff}) for the depletion steps are shown in Fig. 3 and Table 2. The standard deviations of the multiplication factor are within 20 pcm.

Cumulative plutonium production at each depletion step calculated by MCS simulation is presented in Fig. 4. Table 3 shows the plutonium isotopes production calculated by MCS in Calder Hall reactor at burnup steps. Since the other isotopes except for ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu and ^{242}Pu are produced with less than 0.01 kg, the total plutonium production is assumed to be the sum of ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu and ^{242}Pu production. In this study, these values are assumed to be the actual plutonium production from the Calder Hall reactor and they are used as the reference values for the comparison with the values estimated by GIRM combined with polynomial regression in this study.

Table 2. Effective multiplication factors of whole core

Burnup [day]	Whole core	
	k_{eff}	Std. Dev. [pcm]
0.0	1.01661	14
0.2	1.01696	10
0.5	1.01694	20
1.0	1.01666	13
2.0	1.01685	21
5.0	1.01662	13
10.0	1.01665	14
25.0	1.01702	16
50.0	1.01894	15
250.0	1.03309	13
450.0	1.03849	15
650.0	1.04042	17
850.0	1.03998	20
1,050.0	1.03835	19
1,250.0	1.03587	19
1,450.0	1.03249	17
1,650.0	1.02873	16
1,850.0	1.02521	16
2,050.0	1.02074	12
2,250.0	1.01666	17
2,450.0	1.01214	19
2,650.0	1.00724	19
2,850.0	1.00298	16
3,050.0	0.99862	21
3,250.0	0.99403	17
3,450.0	0.98900	20
3,650.0	0.98420	15
3,850.0	0.97942	17
4,050.0	0.97529	20
4,250.0	0.97034	22

2.2 Graphite Isotope Ratio Method (GIRM)

The procedure of GIRM combined with polynomial regression applied in this study is as follow. First, through 2-D unit fuel pin cell depletion calculation, the cumulative plutonium mass density is tabulated as a function of the ^{10}B/^{11}B ratio. Fig. 5 shows the plutonium mass density as

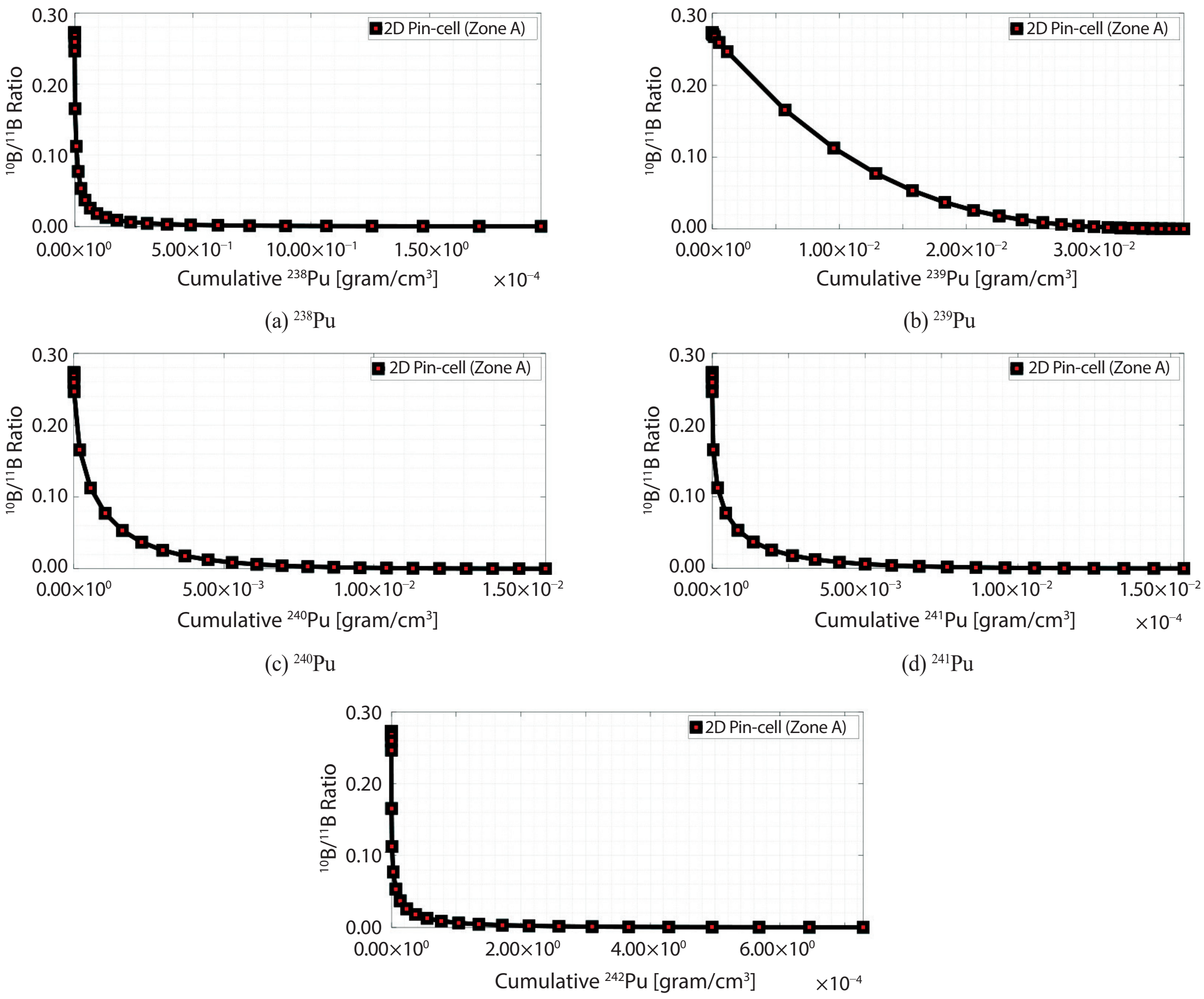
Table 3. Cumulative plutonium isotopes production in Calder Hall reactor at burnup steps

Burnup [day]	Mass [kg]				
	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu
0.0	0.00	0.00	0.00	0.00	0.00
50.0	0.00	8.51	0.07	0.00	0.00
250.0	0.00	40.53	1.59	0.07	0.00
450.0	0.01	66.99	4.47	0.35	0.01
650.0	0.01	89.49	8.23	0.83	0.04
850.0	0.02	109.03	12.59	1.50	0.09
1,050.0	0.04	126.25	17.40	2.30	0.16
1,250.0	0.06	141.58	22.54	3.22	0.27
1,450.0	0.09	155.31	27.96	4.22	0.41
1,650.0	0.12	167.70	33.58	5.30	0.58
1,850.0	0.16	178.95	39.36	6.44	0.78
2,050.0	0.22	189.17	45.28	7.62	1.02
2,250.0	0.28	198.53	51.30	8.85	1.30
2,450.0	0.35	207.03	57.40	10.11	1.61
2,650.0	0.43	214.86	63.56	11.39	1.95
2,850.0	0.53	222.04	69.76	12.69	2.34
3,050.0	0.63	228.63	76.00	14.01	2.75
3,250.0	0.76	234.69	82.25	15.34	3.21
3,450.0	0.89	240.24	88.51	16.68	3.70
3,650.0	1.04	245.38	94.76	18.02	4.23
3,850.0	1.21	250.10	101.00	19.37	4.79
4,050.0	1.39	254.46	107.22	20.71	5.39
4,250.0	1.59	258.46	113.41	22.05	6.03

a function of the $^{10}B/^{11}B$ ratio in 2-D fuel pin. Then, the cumulative plutonium mass density at a sampling point in the reactor is estimated using the $^{10}B/^{11}B$ ratio sampled from the 3-D simulation and the tabulated cumulative plutonium mass density function. Alternatively, $^{10}B/^{11}B$ ratio from the 3-D simulation can be replaced by a measured data in actual application of GIRM. The 3-D spatial distribution of plutonium mass density over the entire core is derived through least-squares regression using the estimated plutonium mass density for each sampling point. Finally, the total plutonium production in the core can be estimated by integrating the 3-D spatial distribution of plutonium mass density over the entire core. The accuracy of GIRM combined with polynomial regression can be evaluated by comparing the total plutonium production estimated by GIRM combined with polynomial regression and that from the 3-D core depletion calculation using MCS.

The radial and axial sampling points in the Calder Hall reactor are given in Fig. 6. Since the configuration of fuel pins has a quarter core symmetry in a whole core, the sampling points were selected within a quarter core. The number of radial and axial sampling points are 28 and 5, respectively. Therefore, a total of 140 sampling data were used.

A 3-D space-dependent least-squares regression function based on the triangular basis was used to calculate the plutonium mass density for the whole core as shown in Eq. (1).



(a) ^{238}Pu

(b) ^{239}Pu

(c) ^{240}Pu

(d) ^{241}Pu

(e) ^{242}Pu

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

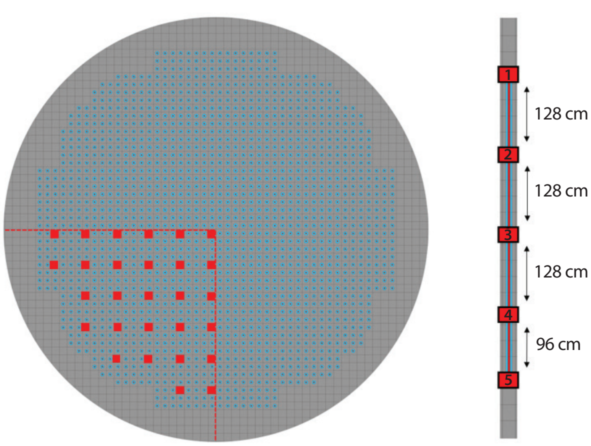


Fig. 6. Radial and axial sampling points.

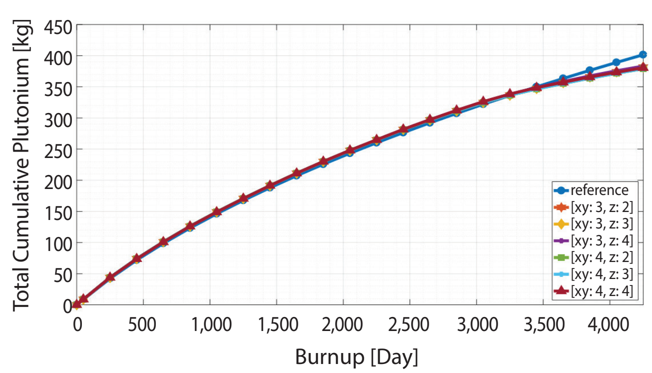


Fig. 7. Comparison of the cumulative plutonium production with different orders of regression.

Table 4. Comparison of total cumulative plutonium production calculated by MCS and GIRM

Burnup [day]	Total Cumulative Plutonium [kg]		Relative Error [%]	R^2 of the Regression
	MCS	GIRM		
0.0	0.000	0.000	0.0	-
50.0	8.578	8.843	3.1	0.9959
250.0	42.194	43.605	3.3	0.9953
450.0	71.819	73.644	2.5	0.9959
650.0	98.606	100.790	2.2	0.9960
850.0	123.234	125.842	2.1	0.9962
1050.0	146.155	148.780	1.8	0.9962
1250.0	167.669	170.519	1.7	0.9962
1450.0	187.984	191.281	1.8	0.9963
1650.0	207.282	211.143	1.9	0.9963
1850.0	225.696	230.015	1.9	0.9964
2050.0	243.317	247.998	1.9	0.9964
2250.0	260.244	264.965	1.8	0.9964
2450.0	276.495	281.749	1.9	0.9964
2650.0	292.190	297.264	1.7	0.9965
2850.0	307.351	312.186	1.6	0.9965
3050.0	322.023	326.069	1.3	0.9964
3250.0	336.239	338.220	0.6	0.9960
3450.0	350.017	348.934	−0.3	0.9949
3650.0	363.432	358.491	−1.4	0.9933
3850.0	376.472	367.635	−2.3	0.9910
4050.0	389.180	375.841	−3.4	0.9892
4250.0	401.551	383.328	−4.5	0.9879

$$f(x,y,z)=\sum_{k=0}^{K}\sum_{\substack{i=0\\j=0}}^{i+j\le N} a_{i,j,k}x^i y^j z^k \tag{1}$$

where $f(x, y, z)$ is the plutonium mass density for the (x, y, z) location in the core, K and N are the regression orders for the axial and the radial directions, respectively. In this study, several orders for the axial and the radial directions were tested as shown in Fig. 7 in order to find an optimized result. The results are similar to each other and quite good accuracy was observed regardless of the polynomial order. It is ascribed to the fact that the core has no control rod and therefore the flux shape is very smooth throughout

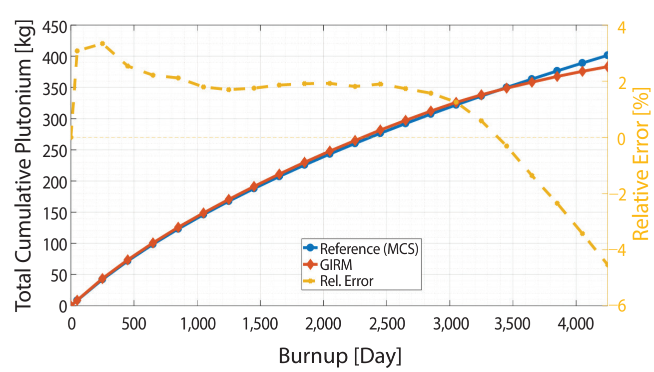


Fig. 8. Comparison of total cumulative plutonium production calculated by MCS and GIRM.

Table 5. Comparison of axial cumulative plutonium production calculated by MCS and GIRM at a burnup step of 1250 day

Height [cm]		Total Cumulative Plutonium [kg]		Relative Error [%]
Bottom	Top	MCS	GIRM	
100	132	5.333	5.774	8.3
132	164	6.282	6.604	5.1
164	196	7.179	7.364	2.6
196	228	7.932	8.046	1.4
228	260	8.564	8.644	0.9
260	292	9.082	9.152	0.8
292	324	9.494	9.564	0.7
324	356	9.806	9.878	0.7
356	388	9.997	10.089	0.9
388	420	10.105	10.196	0.9
420	452	10.108	10.197	0.9
452	484	10.017	10.093	0.8
484	516	9.803	9.884	0.8
516	548	9.502	9.572	0.7
548	580	9.096	9.159	0.7
580	612	8.577	8.649	0.8
612	644	7.947	8.046	1.2
644	676	7.189	7.356	2.3
676	708	6.298	6.585	4.6
708	740	5.357	5.740	7.1

Table 6. Comparison of axial cumulative plutonium production calculated by MCS and GIRM at a burnup step of 2250 day

Height [cm]		Total Cumulative Plutonium [kg]		Relative Error [%]
Bottom	Top	MCS	GIRM	
100	132	8.795	9.540	8.5
132	164	10.246	10.762	5.0
164	196	11.515	11.823	2.7
196	228	12.524	12.732	1.7
228	260	13.349	13.497	1.1
260	292	13.996	14.124	0.9
292	324	14.487	14.619	0.9
324	356	14.856	14.986	0.9
356	388	15.080	15.228	1.0
388	420	15.207	15.348	0.9
420	452	15.205	15.347	0.9
452	484	15.097	15.224	0.8
484	516	14.863	14.978	0.8
516	548	14.506	14.607	0.7
548	580	14.011	14.108	0.7
580	612	13.366	13.475	0.8
612	644	12.552	12.704	1.2
644	676	11.520	11.786	2.3
676	708	10.254	10.715	4.5
708	740	8.816	9.481	7.5

the entire core. Cubic order for radial (K = 3) and quartic order for axial direction (N = 4) was chosen for the rest part of this study.

3. Results

3.1 Whole Core Plutonium Production

The total cumulative plutonium production calculated by MCS and that estimated by GIRM combined with polynomial regression were compared at each burnup steps as shown in Table 4 and Fig. 8. The maximum error of 3.1% is observed at the first burnup step and the error at the last burnup step is −4.5%. The root mean squares (RMS) of the errors throughout the burnup steps is 2.2%. The coefficients of determination (R2) for polynomial regression are also given for all depletion steps in Table 4. They are close enough to 1.0 (between 0.9879 and 0.9965), which indicates that the polynomial regression function was derived properly.

3.2 Axial and Pin-wise Plutonium Production

To verify the accuracy of polynomial regression, the plutonium productions at various axial points and those at various fuel pins were compared at the burnup steps of 1250, 2250, and 3250 days. Table 5–7 compare the axial cumulative plutonium production calculated by MCS and estimated by GIRM combined with polynomial regression

Table 7. Comparison of axial cumulative plutonium production calculated by MCS and GIRM at a burnup of 3250 day

Height [cm]		Total Cumulative Plutonium [kg]		Relative Error [%]
Bottom	Top	MCS	GIRM	
100	132	11.926	12.935	8.5
132	164	13.762	14.540	5.7
164	196	15.261	15.778	3.4
196	228	16.405	16.715	1.9
228	260	17.292	17.409	0.7
260	292	17.964	17.911	−0.3
292	324	18.460	18.261	−1.1
324	356	18.817	18.494	−1.7
356	388	19.032	18.634	−2.1
388	420	19.153	18.698	−2.4
420	452	19.155	18.694	−2.4
452	484	19.051	18.622	−2.3
484	516	18.819	18.475	−1.8
516	548	18.475	18.235	−1.3
548	580	17.975	17.878	−0.5
580	612	17.306	17.369	0.4
612	644	16.421	16.668	1.5
644	676	15.266	15.725	3.0
676	708	13.767	14.481	5.2
708	740	11.930	12.869	7.9

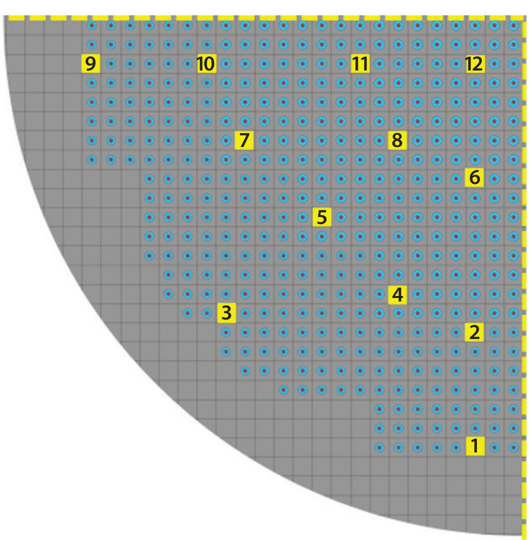


Fig. 9. Fuel pin index for pin-wise comparison of plutonium production.

Table 8. Comparison of pin-wise cumulative plutonium production calculated by MCS and GIRM at a burnup step of 1250 day

Fuel Pin Index	Total Cumulative Plutonium [kg]		Relative Error [%]
	MCS	GIRM	
1	0.050	0.047	−4.9
2	0.101	0.106	5.0
3	0.057	0.058	2.9
4	0.105	0.108	3.1
5	0.110	0.113	2.7
6	0.134	0.138	2.6
7	0.103	0.106	2.2
8	0.134	0.135	0.7
9	0.049	0.049	−1.5
10	0.100	0.101	0.7
11	0.135	0.135	0.3
12	0.146	0.145	−0.7

Table 9. Comparison of pin-wise cumulative plutonium production calculated by MCS and GIRM at a burnup step of 2250 day

Fuel Pin Index	Total Cumulative Plutonium [kg]		Relative Error [%]
	MCS	GIRM	
1	0.086	0.087	0.2
2	0.159	0.165	4.1
3	0.098	0.101	4.0
4	0.163	0.169	3.5
5	0.171	0.175	2.8
6	0.197	0.201	1.7
7	0.163	0.166	1.6
8	0.196	0.198	1.0
9	0.086	0.089	3.0
10	0.158	0.159	0.6
11	0.197	0.198	0.3
12	0.208	0.206	−0.9

at each burnup step. Although relatively large errors are observed at the top and bottom regions of active core, the RMS errors are 3.1%, 3.2%, and 3.5% at depletion steps of 1250, 2250, and 3250 days respectively. The correlation between the cumulative plutonium mass density and $^{10}B/^{11}B$ ratio is calculated based on a 2-D unit fuel pin cell. It has somewhat similar conditions along the central region of the

Table 10. Comparison of pin-wise cumulative plutonium production calculated by MCS and GIRM at a burnup step of 3250 day

Fuel Pin Index	Total Cumulative Plutonium [kg]		Relative Error [%]
	MCS	GIRM	
1	0.121	0.125	3.0
2	0.206	0.212	2.8
3	0.136	0.142	4.0
4	0.211	0.217	2.7
5	0.219	0.225	2.4
6	0.242	0.240	−0.6
7	0.212	0.215	1.4
8	0.242	0.239	−1.3
9	0.121	0.125	3.7
10	0.207	0.208	0.3
11	0.243	0.238	−1.9
12	0.251	0.238	−5.0

core, but not in the top and bottom regions, where the spectrum is different because of the axial reflectors. Therefore, the top and bottom active core regions show a relatively larger error.

As for the pin-wise cumulative plutonium production, several fuel pin locations in different regions of the quarter core were selected. The chosen locations are shown in Fig. 9. The comparison of cumulative plutonium production calculated using both MCS and GIRM for each chosen fuel pin is presented in Table 8–10. The relative errors were found within acceptable range.

4. Conclusion

In this study, the accuracy of the GIRM combined with polynomial regression to estimate the total cumulative plutonium production was verified. The cumulative plutonium production in the Calder Hall reactor was estimated by the GIRM combined with polynomial regression and the results were compared with those calculated by a 3-D Monte-Carlo depletion calculation using the MCS code. With cubic and quartic order regression in axial and radial direction, respectively, the RMS error throughout the burnup steps is about 2.2%. Although the errors at top and bottom of the active core are relatively large, the error of the regression with cubic and quartic polynomial in axial and radial direction was acceptable. The RMS error was around 3.3%. The accuracy of the cumulative plutonium production estimated by the GIRM combined with polynomial regression at 12 fuel pins also assessed. The RMS error was about 2.4–2.8% depending on the burnup steps.

It was found that the accuracy of the regression with cubic and quartic polynomial was satisfactory for the estimation of cumulative plutonium production in Calder Hall reactor. However, no control rod was considered during the reactor operation in this study. The use of control rod will cause a distortion of flux shape. In that case, the accuracy of the cubic and quartic polynomial regression could be doubtful. The effect of the control rod on the accuracy of the polynomial regression are expected to be assessed in further works.

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