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Application of Graphite Isotope Ratio Method with Artificial Intelligence to Estimate Cumulative Plutonium Production in a Graphite-Moderated Reactor

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1. Introduction

The international communities, including the United States of America and the Republic of Korea, have been trying to denuclearize the Democratic People's Republic of Korea (DPRK). If DPRK implements an agreement on non-proliferation of nuclear weapons, it is essential to estimate the total plutonium production and to verify the plutonium production declared by DPRK [1,2]. A Magnox-type reactor at Yongbyon Nuclear Scientific Research Center is known to produce weapon-grade plutonium [1]. The total plutonium production of the reactor at Yongbyon could be easily predicted if the reactor information, such as core designs and operating history, should be available. However, additional measurements are essential since the information related to nuclear research in DPRK is confidential [2]. Without detailed information on the reactor, Graphite Isotope Ratio Method (GIRM) can be used to predict the total plutonium production. The concept of GIRM is that the cumulative plutonium production is proportional to the change of the impurity isotope ratio caused by transmutation [3].

Still, it is unable for GIRM to be applied to all fuel channels because the isotope ratio data for each region are limited. In a previous work by Kim *et al.* [1], a 3D Least Square Regression (LSR) method was used to compensate for those lacking data. In the 3D LSR method, however, only 3 inputs were used: (x, y, z) location. The process to determine the optimal orders of z and xy was necessary for nonlinear regression as well. AI regression techniques can easily perform nonlinear regression using more inputs, possibly resulting in higher accuracy. This study presents application of GIRM with Artificial Intelligence (AI) to estimate cumulative plutonium production in a graphite-moderated nuclear reactor. Nonlinear regression can be easily performed using AI.

2. Methods

This section explains the magnox reactor, a reference reactor for prediction, and techniques used to estimate ^{239}Pu production. The techniques include the process of GIRM, estimation process using GIRM with AI, and the parameters of AI.

2.1 Magnox Reactor

The Magnox reactor uses natural uranium as a fuel, graphite as a moderator, and carbon dioxide gas as a heat exchange coolant. It is a smaller version of Britain's Calder Hall reactor, producing the major amount of weapon-grade plutonium in DPRK [4]. The radial layout and design parameters of the reactor are illustrated in Fig. 1 and Table I, respectively.

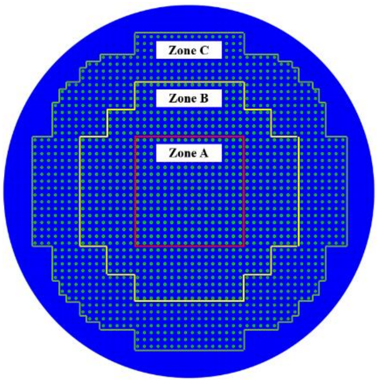

Fig. 1. Radial layout of Magnox reactor [1].

Table. I: Design parameters of Magnox reactor [1].

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

2.2 GIRM

GIRM is a plutonium production verification method that consists of three main steps: the first step to identify the core components containing the most reliable sources of information on the reactor, the step to identify elements in the graphite suitable as indicator element, and the full-scale reactor application step [5]. In the study by Kim *et al.* [1], ^{10}B and ^{11}B were selected as indicator elements, and ^{10}B/^{11}B isotopic ratio was used to calculate plutonium production. Fig. 2 shows the plutonium mass density for the boron isotopic ratio in a 2D fuel pin. The total estimated ^{239}Pu production was calculated by integrating the equation derived through 3D LSR over the whole core region.

2.3 GIRM with AI

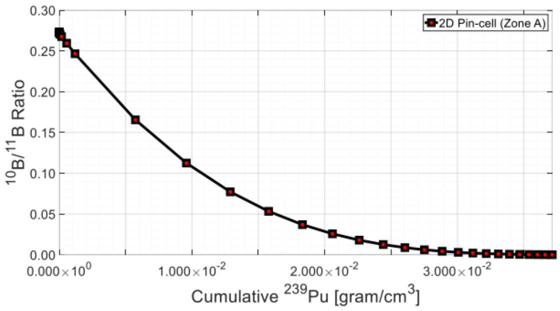

Fig. 2. ^{239}Pu mass density for ^{10}B/^{11}B ratio in a 2D fuel pin [1].

This study estimated the total cumulative ^{239}Pu production using AI regression. First, ^{239}Pu data in gram per cubic centimeter at the sampling regions of the Magnox reactor were given as training data. These data have been used initially in work by Kim *et al.* [1]. Fig. 3 illustrates the axial and radial sampling regions of the quarter core. There are 140 sampling regions, with 28 radial and five axial regions. The next step was to preprocess the data so that all data were normalized in the range of (0, 1).

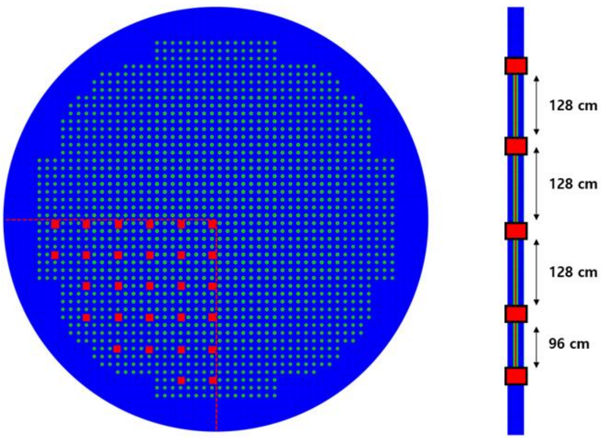

Fig. 3. The axial and radial sampling region [1].

The cumulative ^{239}Pu mass was then calculated as follows. With the *x*, *y*, and *z* index of each sampling region, the corresponding burnup as inputs, and the reference ^{239}Pu data calculated by MCS code as a label, the designed AI is trained. For every *x* and *y* index of the fuel pin in the quarter core, ^{239}Pu production data in gram per cubic centimeter at *z* indices (1, 2, 3, …, 19, 20) are predicted. Multiplying these values by the volume of one axial point (approximately 214.585 cubic centimeters) accounts for the produced ^{239}Pu in gram at the point. Adding up these values in the single channel gives the cumulative ^{239}Pu production at that fuel channel. Finally, the cumulative ^{239}Pu production in the quarter core was calculated by adding up the cumulative ^{239}Pu production in every fuel channel. Additionally, the axial cumulative ^{239}Pu production was calculated as well by adding up the values in the regions with the same *z* indices.

2.4 AI Model

A Multi-Layer Perceptron (MLP) is selected as a deep learning model to solve the AI regression task. The AI regression model for the estimation of ^{239}Pu is designed using Keras, an open-source Python deep learning API. The model consists of 4 input nodes corresponding to the region's *x*, *y*, *z* index, and the burnup, respectively, and 1 output node corresponding to ^{239}Pu density. The total number of the data was 4200, 140 sampling regions per burnup for 30 burnup steps. 5% of the data were divided as a validation dataset using validation_split parameter in Keras. The total epochs were 600, the batch size was 32 (default), the optimizer was Adam with a learning rate of 0.0001, and the loss function was Mean Squared Error (MSE). The parameters used to train the MLP were listed in Table. II.

Table. II: Parameters for training MLP.

Parameter	Value
Total number of data	4200
Number of validation data	210 (5% of total data)
Total number of epochs	600
Batch size	32 (default)
Optimizer	Adam
Learning rate	0.0001
Loss function	Mean squared error

3. Results

The Absolute Percentage Error (APE) is used for the evaluation. The formula for APE is given as follows:

$$\text{APE (\%)} = \left|\frac{P_{ref}-P_{pred}}{P_{ref}}\right| * 100, \quad (1)$$

where P_{ref} corresponds to total plutonium production calculated by MCS and P_{pred} to the production predicted by the MLP. The calculation results of the whole core, axial, and pin-wise calculations and APEs are presented in each subsection below.

3.1 Whole Core Calculation

Table. III and Fig. 4 show the estimated cumulative ^{239}Pu production in the whole core. The estimation using LSR has 1.339% APE on average. The estimation using AI has 0.860% APE on average.

Table. III: Total cumulative ^{239}Pu production results calculated by MCS, LSR, and MLP.

Burnup [day]	Total ^{239}Pu production [kg]			Error [%]	
	MCS	LSR	MLP	LSR	MLP
50	8.51	8.77	8.74	3.036	2.744

250	40.53	41.07	40.53	1.336	0.003
450	66.99	68.09	67.33	1.653	0.522
650	89.49	90.98	89.83	1.657	0.378
850	109.03	110.82	109.56	1.643	0.486
1050	126.25	128.10	126.67	1.461	0.331
1250	141.58	143.54	142.09	1.384	0.365
1450	155.31	157.55	156.09	1.441	0.501
1650	167.70	170.40	168.96	1.609	0.752
1850	178.94	182.07	180.78	1.749	1.027
2050	189.17	192.61	191.53	1.816	1.245
2250	198.53	201.99	201.19	1.748	1.341
2450	207.03	210.62	209.82	1.734	1.347
2650	214.86	218.07	217.49	1.494	1.226
2850	222.04	224.81	224.28	1.249	1.010
3050	228.63	230.86	230.31	0.975	0.736
3250	234.68	236.16	235.62	0.629	0.397
3450	240.24	240.94	240.27	0.291	0.013
3650	245.38	245.20	244.36	0.075	0.416
3850	250.10	249.08	247.92	0.410	0.872
4050	254.46	252.40	251.01	0.812	1.357
4250	258.46	255.23	253.68	1.250	1.849

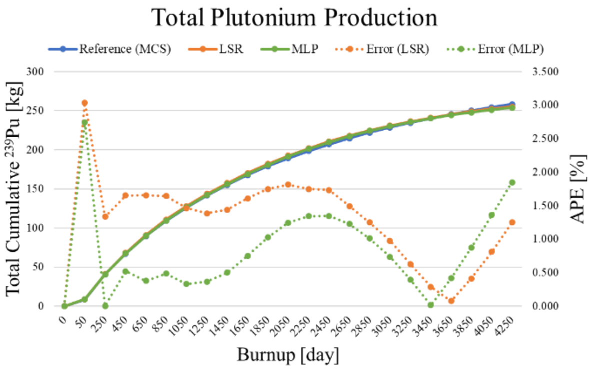

Fig. 4. Total cumulative ^{239}Pu production results calculated by MCS, LSR, and MLP.

3.2 Axial Calculation

Table. IV and Fig. 5 show the estimated cumulative ^{239}Pu production in the axial regions. The estimation using LSR has 1.687% APE on average. The estimation using AI has 1.629% APE on average.

Table. IV: Axial cumulative ^{239}Pu production results on day 3250 calculated by MCS, LSR, and MLP.

Bottom ~ Top [cm]	Axial ^{239}Pu production [kg]			Error [%]	
	MCS	LSR	MLP	LSR	MLP
100~132	9.208	10.047	9.981	9.112	8.393
132~164	10.421	10.586	10.748	1.583	3.135
164~196	11.174	11.069	11.343	0.940	1.514
196~228	11.682	11.496	11.760	1.592	0.671
228~260	12.045	11.865	12.044	1.494	0.011
260~292	12.301	12.178	12.249	1.000	0.419
292~324	12.477	12.431	12.407	0.369	0.565
324~356	12.607	12.625	12.525	0.143	0.652
356~388	12.681	12.759	12.586	0.615	0.750
388~420	12.719	12.832	12.596	0.888	0.969
420~452	12.725	12.843	12.563	0.927	1.270
452~484	12.689	12.791	12.508	0.804	1.424
484~516	12.606	12.676	12.439	0.555	1.325
516~548	12.485	12.497	12.355	0.096	1.044
548~580	12.309	12.253	12.229	0.455	0.649
580~612	12.056	11.943	12.004	0.937	0.433
612~644	11.691	11.566	11.642	1.069	0.423
644~676	11.176	11.122	11.170	0.483	0.050
676~708	10.423	10.609	10.577	1.785	1.482
708~740	9.209	10.028	9.891	8.893	7.401

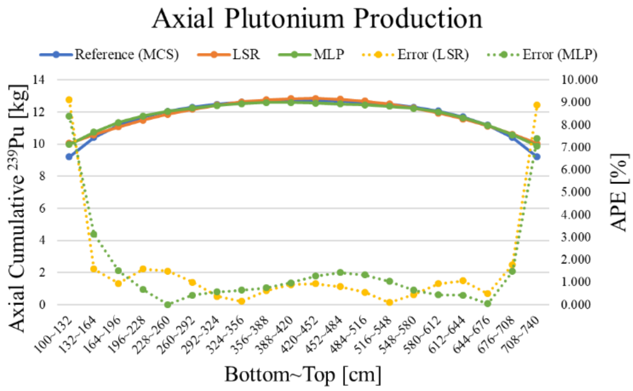

Fig. 5. Axial cumulative ^{239}Pu production results on day 3250 calculated by MCS, LSR, and MLP.

3.3 Pin-Wise Calculation

Several fuel pins, illustrated in Fig. 6, were selected in work by Kim *et al.* [1] for the calculation and comparison of pin-wise production. Table. V and Fig. 7 show the estimated ^{239}Pu production in the selected fuel pins. The estimation using LSR has 2.016% APE on average. The estimation using AI has 2.203% APE on average.

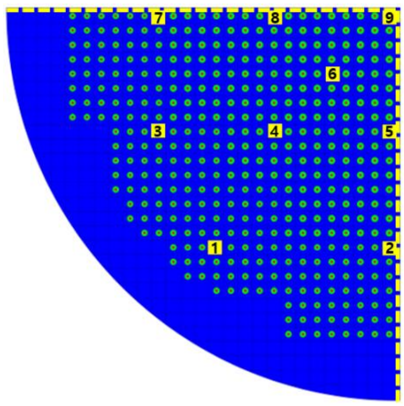

Fig. 6. Fuel pins and their indices for pin-wise comparison of ^{239}Pu production [1].

Table. V: Pin-wise cumulative ^{239}Pu production results on day 3250 calculated by MCS, LSR, and MLP.

Pin Index	Pin-wise ^{239}Pu production [kg]			Error [%]	
	MCS	LSR	MLP	LSR	MLP
1	0.121	0.124	0.124	2.479	2.108
2	0.145	0.148	0.148	2.069	2.412
3	0.135	0.137	0.138	1.481	2.252
4	0.155	0.154	0.152	0.645	2.151
5	0.157	0.155	0.154	1.274	1.665
6	0.158	0.153	0.154	3.165	2.248
7	0.144	0.146	0.146	1.389	1.292

8	0.158	0.154	0.154	2.532	2.458
9	0.161	0.156	0.156	3.106	3.245

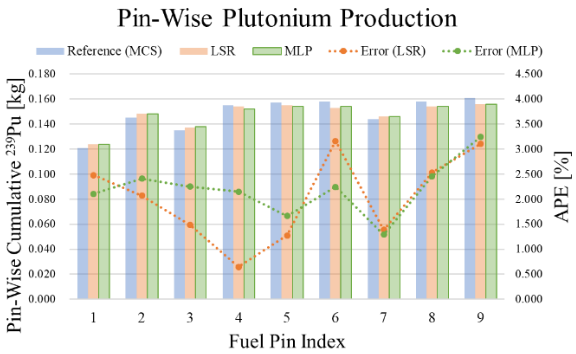

Fig. 7. Pin-wise cumulative ^{239}Pu production results on day 3250 calculated by MCS, LSR, and MLP.

4. Conclusions

Further to work by Kim *et al* [1], the cumulative plutonium production in the Magnox reactor was predicted using AI, and the result was compared to those calculated by MCS and LSR. AI shows more inaccurate result in the pin-wise calculation, with 0.187% point larger APE. The prediction using AI has larger APE in the burnups greater than or equal to 3650. Nevertheless, the estimation by AI shows higher overall accuracy, with 0.479 and 0.058% point lower APEs in whole core and axial calculation, respectively.

Future research will predict the productions of the other isotopes such as ^{240}Pu, ^{242}Pu, and ^{241}Am by adding output nodes of the model. Furthermore, instead of using validation_split, more sampling regions as validation and test dataset will be added in the model training process. This will allow future research to compare plutonium data in certain points instead of whole core data as in this work.

REFERENCES

[1] Kyeongwon Kim, Jinseok Han, Ran Lee, Hyun Chul Lee, Junkyung Jang, and Deokjung Lee, "Estimation of Plutonium Production in a Graphite-moderated Reactor Using Graphite Isotope Ratio Method and MCS", Transactions of the Korean Nuclear Society Spring Meeting, May 2021.
[2] P. G. Heasler, T. Burr, B. Reid, C. Gesh, and C. Bayne, "Estimation Procedures and Error Analysis for Inferring the Total Plutonium (Pu) Produced by a Graphite-Moderated Reactor", Reliability Engineering and System Safety, Vol. 91, pp. 1406-1413, 2006, DOI: 10.1016/j.ress.2005.11.036.
[3] Jungmin Kang, "Using the Graphite Isotope Ratio Method to Verify the DPRK's Plutonium-Production Declaration", Science and Global Security, Vol. 19, pp. 121-129, 2011, DOI: 10.1080/08929882.2011.586309.
[4] C. Braun, S. Hecker, C. Lawrence, P. Papadiamantis, "North Korean Nuclear Facilities After the Agreed Framework", Center for International Security and Cooperation, Stanford University, May 2016.
[5] J. P. McNeece, B. D. Reid, and T. W. Wood, "The Graphite Isotope Ratio Method (GIRM): A Plutonium Production Verification Tool", United States, 1999, DOI:10.2172/9672