Improvement of Estimating Plutonium Production using Various Estimation Models for Graphite Isotope Ratio Method

Jinseok Han [a], Kyeongwon Kim [b], Seongjin Jeong [a], Junkyung Jang [a], Hyun Chul Lee [a*]
[a]*School of Mechanical Engineering, Pusan National University, 2, Busandaehak-ro 63beon-gil, Geumjung-gu, Busan, 46241, Republic of Korea*
[b]*Department of Nuclear Engineering, Ulsan National Institute of Science and Technology, 50 UNIST-gil, Ulsan, 44919, Republic of Korea*
[*]*Corresponding author: hyunchul.lee@pusan.ac.kr*

1. Introduction

Since July 2021, indications that the 5MW$_e$ reactor in Yongbyon was operating have been observed. Plutonium is produced at graphite moderated reactor in Yongbyon to make nuclear weapons by the DPRK [1]. For the peaceful use of atomic energy and national security, it is important to denuclearize the DPRK. Accurate prediction of the number of nuclear weapons made with plutonium that North Korea has is necessary for non-proliferation. Graphite Isotope Ratio Method (GIRM) can be used to estimate plutonium produced in graphite moderated reactors. This method was developed by PNNL in the 1990s [2]. GIRM can estimate the amount of plutonium produced in the reactor by investigating the isotope ratio of indicator nuclides in graphite without relying on the operation history of the reactor.

In our previous studies, the selection of suitable indicator nuclides and verification of GIRM through 3D polynomial regression were conducted [3,4]. In this study, the prediction accuracy of GIRM is analyzed when neutron flux fluctuates by inserting control rods during depletion calculation of Magnox reactor. Additionally, the temperature distribution of fuel and graphite inside the reactor was also considered. Boron, titanium, tungsten, and uranium were used as indicator nuclides. Various prediction models, such as 2D pin cells and 3D channels, 3D pans, were used to reduce the inferring error of produced plutonium.

2. Methods

Instead of sampling graphite from an actual reactor core, MCS Monte Carlo code which is developed by Ulsan National Institute of Science and Technology was used to conduct depletion simulation of Magnox reactor for acquisition of sampling data and reference data.

Isotope ratio data of graphite from depletion calculation of the reactor is sampled across the core to predict the amount of produced plutonium at each sampling region. Through the correlation between the indicator isotope ratio and cumulative plutonium from the estimation model, locally estimated plutonium data is used for 3D polynomial regression to derive a function which predicts cumulative plutonium across the core.

2.1 Magnox Reactor

Built by the UK, Magnox reactor is used for both commercial power generation and production of weapon-grade plutonium. Calder Hall is selected in this study among Magnox reactors. Natural Uranium, Graphite and CO_2 are used as fuel, moderator, and coolant. Control rods are made of 18/8 stainless steel containing 4% of Boron [5]. Detailed design parameter is described in Table. 1 below.

Table I: Design parameters of Magnox reactor [5,6]

Parameter		Value
Power		182 MW$_{th}$
Active height		640 cm
Fuel pin radius		1.4610 cm
Cladding radius		2.0400 cm
Coolant radius	Zone A	5.2080 cm
	Zone B	5.0165 cm
	Zone C	4.5847 cm
Control rod radius		3.87 cm
Control rod hole radius		4.125 cm
Average Fuel Temperature		425 ºC
Average Graphite Temperature		250 ºC
Number of fuel channels		1696 EA
Number of Control rods		40 EA
Mass of Uranium		120 tones

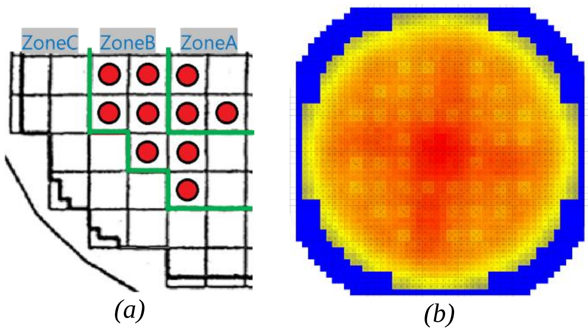


Fig. 1. Radial view of Magnox reactor (a) quadrant core, location of control rod and zone region, (b) radial power distribution at BOC

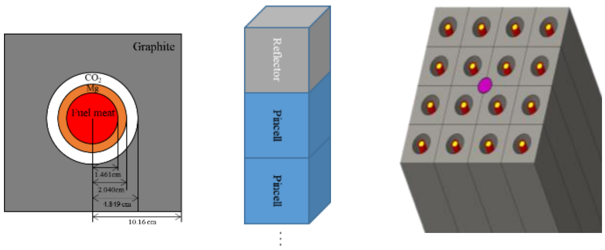
Temperature distribution of the moderator is calculated in proportion to the axial power distribution by zone of the homogeneous core. During criticality calculation, effective multiplication factor is kept at 1 within a range of 40pcm. All control rods were inserted simultaneously with maximum depth 393cm until 650day and withdrawn after. Figure 1 shows radial power distribution at BOC. Depression of power near control rods is observed.

2.2 Estimation Models

Single 2D pin cell model was used in the previous study. 2D pin cell models with different geometry by zone are concerned in this study. Each geometry of 2D pin-cell model is divided into 3 cases which of each is mean temperature of the core, 100K increased and 100K decreased from mean temperature. Coolant radius difference by 3 zones is also concerned.

3D channel model consists of core-height fuel pin cell divided into 20 blocks in the middle and graphite reflectors attached at both axial top and bottom of the fuel pin. Axial temperature distribution of each channel in different zone is considered.

3D pan model has 16(4x4) channels and one control rod hole in the center. Upon features of 3D channel model, pan model is divided into two cases depending on the control rod. One is with control rod that keeps effective multiplication factor at 1. The other case is without control rod. Three different geometries of estimation models are shown at Figure 2.



(a) 2D pin-cell *(b) 3D channel* *(c) 3D pan*

Fig. 2. Geometry of estimation models.

2.3 Local GIRM of sampling data

Sampling of graphite data was carried out at regular intervals in the axial and radial directions for the quadrant core. To make the effect of control rod as low as possible, fuel channel farthest from the control rod is selected. In the axial direction of the selected channel, 5 sampling locations consist of two regions each at the top and bottom moderator of the fuel pin, and three regions in the middle of the channel. In a quadrant core, 140 locations are sampled for 28 channels to derive a function for estimation of produced plutonium by location.

The correlation curves between the isotope ratio and the cumulative plutonium are obtained through depletion simulation of the estimation model. By substituting the sampled isotope ratio data into the correlation curve, the amount of plutonium produced is predicted. The 2D single pin-cell model has one curve per an indicator nuclide.

Number of available correlation curve increases as case of 2D pin-cell model increases. Each geometry of 2D pin-cell models has 3 temperature cases, and each case has its correlation curve that predicts Pu production for input isotope ratio. Plutonium production is interpolated with temperature of sample region.

In case of 3D model, correlation curve at axial location of selected zone can directly substitute sample isotope ratio data into Pu production. Additionally for 3D pan, correlation curve is applied considering relative location from control rod hole and case with or without control rod.

2.4 Global GIRM using 3D Polynomial Regression

3D polynomial regression method using locally estimated plutonium of sampling data is used to estimate global plutonium production of the reactor. Least squares method is applied on the Eq. (1)[4].

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

Function f(x,y,z) represents the mass density of plutonium at 3-D coordinates. Radial location is expressed by x, y coordinates and z coordinates for axial location. Regression orders (N, K) for xy and z decide a regression shape.

Figure 3 shows reference cumulative Pu of the quadrant core with 2850 effective full power days (EFPD) of operation. Each subfigure indicates radial distribution of Pu at certain axial location. Figure 4 shows estimated cumulative Pu using global GIRM.

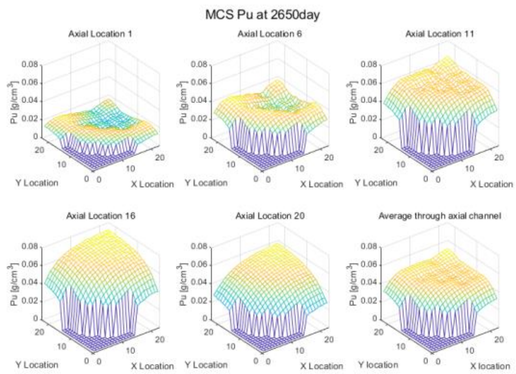


Fig. 3. Cumulative Pu of MCS refference at 2850 day

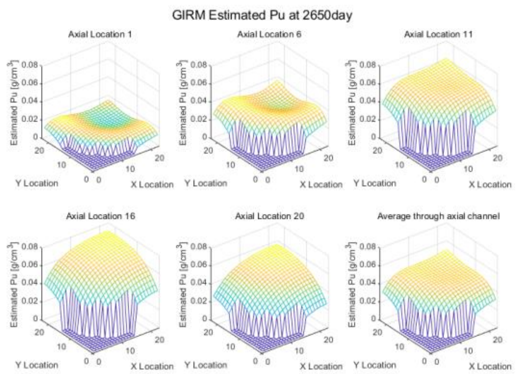


Fig. 4. Cumulative Pu using GIRM at 2850 day

3. Results

In this section, prediction accuracy of estimation model and by indicator is compared through the result of local and global GIRM.

3.1 Local GIRM by estimation models

Boron, Titanium, Tungsten, and Uranium are compared as indicator nuclides. Relative root mean square error(rRMSE) is used to evaluate accuracy of produced Pu of sampling data using GIRM. Figure 5 shows rRMSE of each indicator by estimation model during 50~2850 day of full power operation.

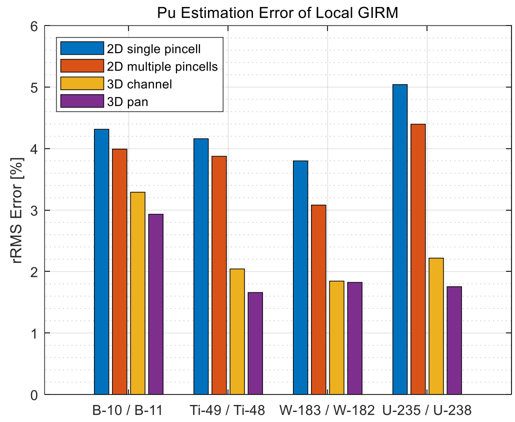


Fig. 5. Pu estimation error using local GIRM.

2D single pin cell model has highest rRMSE between 3.79% and 5.04%. 2D multiple pin cell model shows improvement by value of 3.08% ~ 4.39%. 3D channel and 3D pan models have lowest local estimation error in range of 1.65% ~ 3.29%.

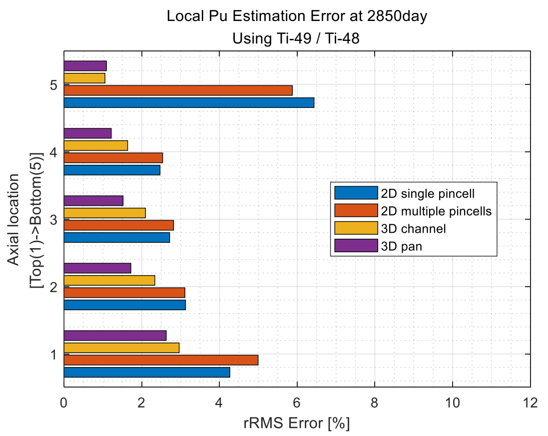


Fig. 6. Axial Pu estimation error using local GIRM with Titanium indicator at 2850day.

When comparing 2D model and 3D model, difference on axial error is remarkable. An example of axial Local GIRM error using Titanium as an indicator is presented on Figure 6. 2D model has higher axial error at both top and bottom region of the core. 3D model shows 4% less error on the bottom region but still has highest error at the top region.

3.2 Global GIRM by estimation models

The coefficient of determination(R^2) is applied to evaluate compatibility of global GIRM using regression method. The regression order for xy and z is decided as 4th order according to the result of Figure 7. Flux fluctuation due to control rod makes optimal regression order higher than previous research which selects 3rd and 4th order each for radial and axial direction. R^2 shows similar results regardless of the estimation model and indicator nuclides.

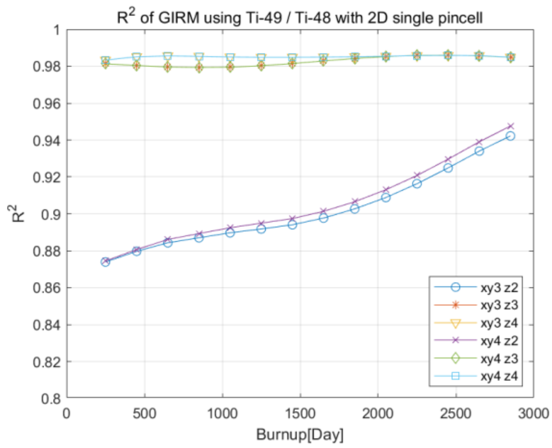


Fig. 7. The coefficients of determination for global GIRM with Ti indicator and 2D single pin cell model.

With 4th regression order for axial and radial direction, comparison of estimation model for global GIRM is conducted. Figure 8 shows estimation of total cumulative plutonium production of each model compared to the MCS reference. Maximum estimation error is found as -2.7% by 3D channel model at 250day. 2D single pin cell model shows the smallest maximum error -1.2% at the same day.

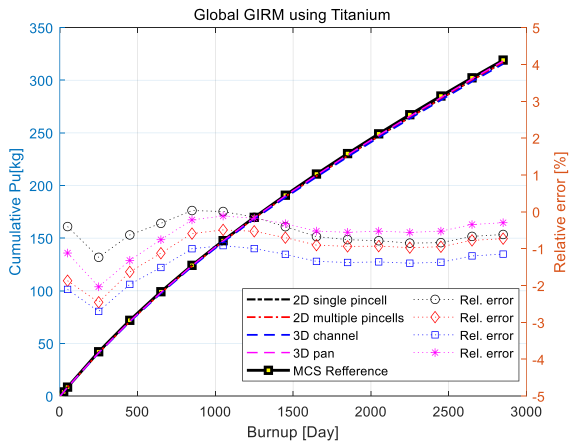


Fig. 8. Estimation of cumulative Pu with Global GIRM using Titanium

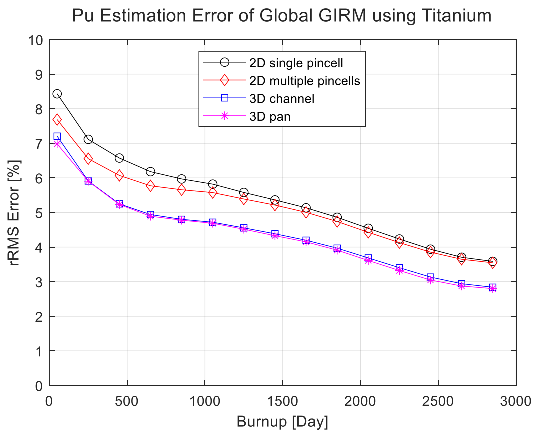


Fig. 9. Pu estimation error using Global GIRM with Titanium indicator.

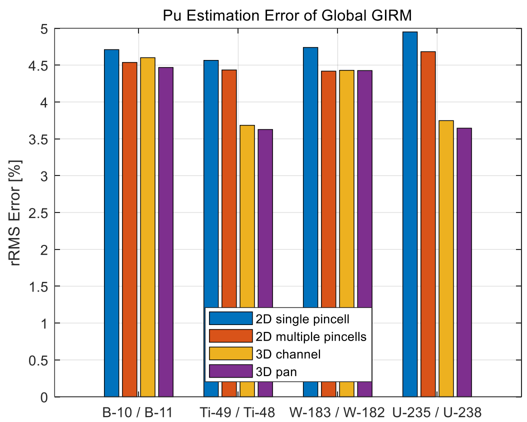


Fig. 10. Pu estimation error using global GIRM.

Even relative error of total plutonium production is low, superposition of error over core could subtract each other and possibly make error smaller. Figure 9 presents rRMSE from global GIRM with titanium indicator. Contrary to the result in relative error of total plutonium, 2D single pin cell model shows biggest error over the whole period. 2D multiple pin cells have slightly better results than 2D single pin. 3D channel and pan have similar error behavior.

Plutonium estimation error using global GIRM in rRMSE for whole burnup period is shown in Fig. 10. In every case, global GIRM with 2D single model has biggest errors. With boron and tungsten indicators, the error difference between 3D models and 2D multiple pin cells is not notable. Contrary, titanium and uranium indicator show 10~20% decrease of rRMSE approximately when using 3D models compared to the 2D models.

4. Conclusions

The overall estimation error of Local GIRM is less when using 3D model than 2D model. Consideration of the reflector effect of the axial graphite has a major impact on the improvement of axial errors. Still estimation error at the top of the core has room for improvement.

As a result of 3D polynomial regression of Global GIRM, rRMSE is flattened compared to the local GIRM. Application of precise estimation model improves plutonium estimation error using global GIRM with titanium and uranium indicator by 10~20% but works poor with boron and tungsten indicator nuclides. Even after considering temperature distribution of the reactor and inserting control rods, GIRM shows plutonium prediction performance within rRMSE less than 5%.

Further evaluation of the prediction performance using GIRM is planned for nuclear fuel reloading in consideration of the optimal operation scenario.

Acknowledgments

This research was supported by the National Research Foundation of Korea(NRF) grant funded by the Korea government(MSIT). (No. NRF-2019R1A2C2089962).

REFERENCES

[1] J. 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
[2] McNeece, J.P., et al., "The Graphite Isotope Ratio Method (GIRM): A Plutonium Production Verification Tool", PNNL-12095, Pacific Northwest National Lab, 1999.
[3] J. Han, J. Jang, and H.C. Lee, "A Suitability Study on the Indicator Isotope for Graphite Isotope Ratio Method (GIRM)", J. Nucl. Fuel Cycle Waste Technol., Vol. 18 No. 1, pp. 83-90, 2020.
[4] K. Kim, J. Han, H.C. Lee, et al., "Verification of Graphite Isotope Ratio Method Combined With Polynomial Regression for the Estimation of Cumulative Plutonium Production in a Graphite-Moderated Reactor", J. Nucl. Fuel Cycle Waste Technol., Vol. 19 No. 4, pp. 447-457, 2021.
[5] Murphy, B. D., "ORIGEN-ARP Cross-Section Libraries for Magnox, Advanced Gas-Cooled, and VVER Reactor Designs", ORNL/TM-2003/263, Oak Ridge National Laboratory, 2004.
[6] S. E. Jensen and E. Nonboel., "Description of the Magnox Type of Gas Cooled Reactor (MAGNOX)", Riso National Laboratory Technical Report, NKS-2, 1999.