Transactions of the Korean Nuclear Society Spring Meeting
Jeju, Korea, May 9-10, 2024

Uncertainty Analysis for Graphite Isotope Ratio Method (GIRM) in a HANARO Simulation

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***Keywords :** Graphite Isotope Ratio Method, estimation of plutonium production, HANARO experimental simulation, estimation of uncertainty, non-proliferation*

1. Introduction

DPRK possesses plutonium produced at a graphite-moderated reactor for weaponization purposes. It poses a significant threat to international security. [1] Therefore, accurately predicting plutonium production becomes crucial to indicate denuclearization progress. The Graphite Isotope Ratio Method (GIRM), developed by PNNL (Pacific Northwest National Lab) in the 1990s, provides a means to predict plutonium quantities by correlating indicator nuclides isotope ratio within graphite impurities, even in cases where detailed operational histories are available. [2-5]

To validate GIRM, the experiments have been conducted at the HANARO, and it is crucial to analyze both errors experimental and simulation errors. This study focuses on investigating errors associated with manufacturing tolerance and simulation. The errors in the simulation stem from both statistical errors caused by random seed numbers and uncertainties arising from covariance in nuclear cross-sections. In this paper, the stochastic errors associated with random seed numbers were evaluated, while errors arising from the uncertainties in nuclear cross-section data will be analyzed in future research. Boron, Iron, Titanium, Tungsten, and Uranium were used as indicator nuclides in previous studies. [3] Regression analysis was conducted to estimate the correlation between the isotope ratios of indicator elements and cumulative plutonium. Additionally, the relative error between predicted and calculated values was analyzed.

2. Methods

For the simulation of HANARO irradiation experiments, the Monte Carlo (MC) code McCARD developed at Seoul National University (SNU) [6] is used. Specimens employed in the experiment include graphite specimens and nuclear fuel specimens. The isotope ratio of indicator elements and cumulative plutonium data required for the GIRM uncertainty evaluation were sampled from depletion calculations performed on the two specimens.

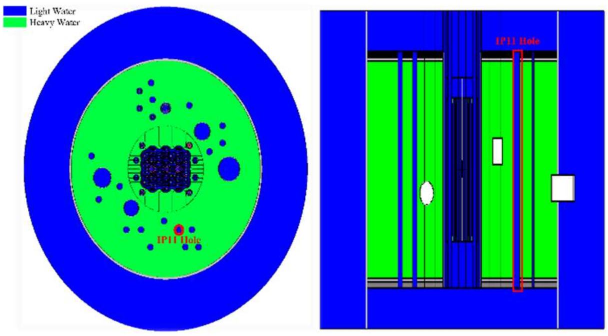
In this paper, uncertainty factors were defined as stochastic error of MC code and manufacturing tolerance of experiment instruments. One hundred independent depletion simulations were conducted for each uncertainty factor. The predicted line using polynomial regression was drawn based on a correlation between the isotope ratio of indicator elements and the amount of plutonium. The uncertainties were estimated through analysis of the distribution of residuals between the predicted and sampled data.

2.1 HANARO Reactor

The HANARO research reactor, open-tank-in pool type, was designed for various purposes such as the production of cold neutrons, irradiation experiments, and others. Uranium silicide is used as nuclear fuel, and its enrichment is 19.75 wt%. Eight Control rods, made of natural hafnium, are used for the operation regulation and shutdown. Heavy water (D_2O) serves as a reflector, while light water (H_2O) is used as a coolant. [7] The detailed parameters of HANARO are described in Table. 1 below.

Table. 1: Design parameters of HANARO [6]

Parameter		Value
Power		30 MW_{th}
Cycle length		28 FPDs
Effective Fuel Length		70 cm
Fuel pin diameter	18-elements	5.486 mm
	36-elements	6.35 mm
Cladding thicknesses	18-elements	1.192 mm
	36-elements	0.76 mm
Control rod diameter		4.5 mm
Max. design temperature of fuel		350 ℃
Inlet coolant temperature		35 ℃
Outlet coolant temperature		44.2 ℃
Number of experimental facilities	Vertical	25
	Tangential	7



(a) (b)

Fig. 1. View of HANARO reactor (a) radial view of a full core, location of IP11 hole, (b) axial view, location of IP11 hole

The neutron irradiation experiments are conducted in an IP11 hole in the reflector region because the size of specimens is small, and parts of specimens are withdrawn during the experiments. Fig.1 shows the position of the IP11 hole, which is the experiment site.

2.2 Experiment instrument specifications

The nuclear specimens used in experiments are UO_2 spheres with a diameter of 0.5mm. The enrichment of nuclear fuel specimens is 0.3 wt%. The graphite specimens are cylinders of equal height and diameter. Each group of four specimens is inserted into a rod. A Jig assembly accommodates three rods, one with a nuclear fuel specimen and the remainder with graphite specimens. The rods are arranged at intervals of 120° on the Jig. The rod inserted nuclear fuel specimen is named Rod1 and is positioned close to the core of the HANARO. The rod named Rod2 is located in the lower-left position, while Rod3 is located in the lower-right position. A Rig assembly contains three Jig assemblies vertically and a Spacer with a similar shape but different heights. The Jig was named Jig1, Jig2, and Jig3 from bottom to top. The Rig assembly is loaded in the IP11 hole for experimental purposes. Therefore, the depletion calculations were performed for 12 nuclear fuel specimens and 24 graphite specimens. The detailed features of the experiment instruments are shown in Fig. 2.

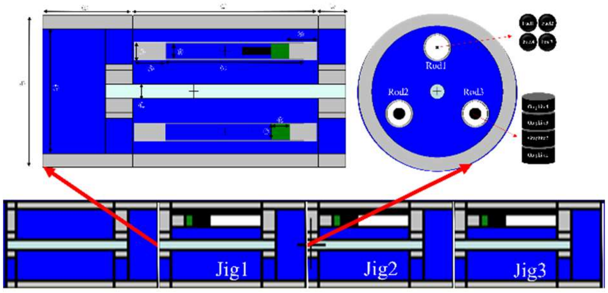


Fig. 2. Experimental instruments (Rig assembly) and internal structure (Jig and rod, specimens) geometry specification, index of tolerance parameters specification.

The dimensions of the rod and Jig have tolerances, and the detailed tolerance parameters are summarized in Table 2.

Table. 2: Specification of experimental manufacturing tolerance

Index	Name	Value [cm]	Tolerance [cm]
1	Jig upper capsule	1.5	±0.04
2	Jig tube	10.8	±0.03
3	Jig under capsule	4.8	±0.02
4	Jig outer radius	2.7	+0/-0.0025
5	Jig inner radius	2.235	+0.005/-0
6	Rod upper capsule	1.7	±0.02
7	Rod tube	7.6	+/-0.03
8	Rod under capsule	1.5	+/-0.02
9	Rod outer radius	0.475	+0/-0.005
10	Rod inner radius	0.4175	+0.001/-0
11	Socket radius	0.25	+/-0.005
12	Insulation radius	0.4095	+/-0.0005
13	Insulation length	1.0	+/-0.001

2.3 Estimation model and sampling data

Due to the small dimensions of graphite and nuclear fuel specimens, it was difficult to accurately calculate the results due to the large relative errors in tally data, such as the flux and reaction rate of the specimens. Hence, the variance reduction technique was essential for reducing relative error. The IMP option of the McCARD code, which conducts geometrical splitting and Russian roulette, was employed. A variance reduction model was designed to split particles as they approach the IP11 hole and specimens in the Jig. Fig. 3 illustrates a radial image of the geometry split model. As particles pass through the cell surface in the direction indicated by the red arrow, they are split. Conversely, in the opposite direction, they gradually disappear based on the IMP option.

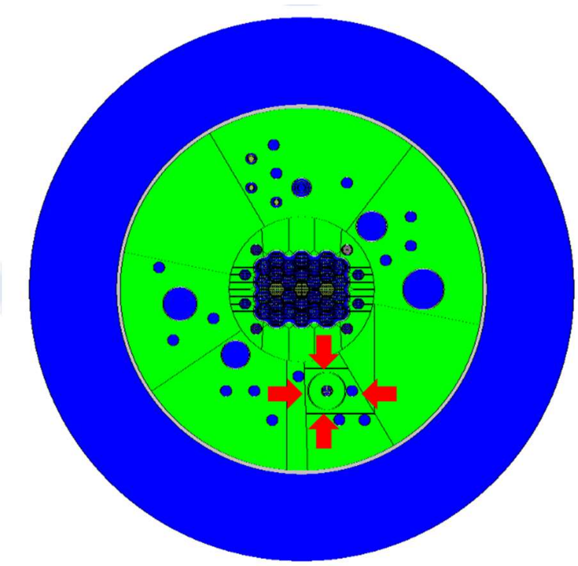


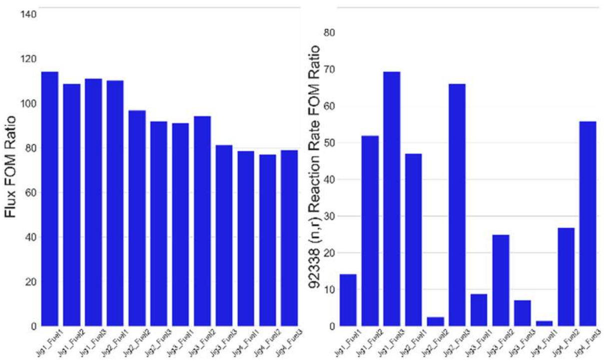
Fig. 3. View of HANARO with geometrical variance reduction model image

The implementation of the geometric variance reduction method increased in computational time. Consequently, it became crucial to determine the appropriate number of particles and the selection of optimal IMP options became imperative. To evaluate the performance of the model with variance reduction, a Figure of Merit (FOM) was employed, as shown in Eq. (1)

$$FOM = \frac{1}{R^2 T} \tag{1}$$

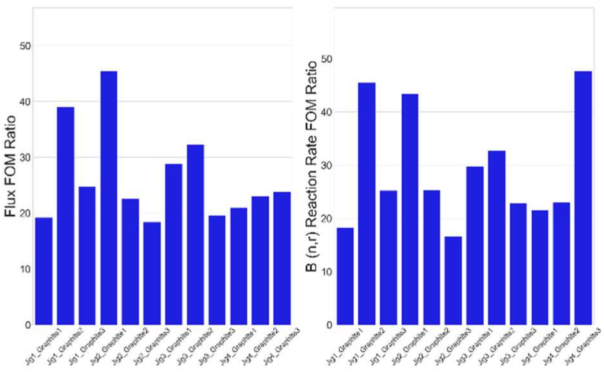
R represents the relative error of parameters tallied from McCARD and, T denotes the computation time spent by processes. An evaluation was conducted on parameters such as the flux and ^{238}U (n,γ) reaction rate of nuclear fuel specimens, as well as the flux and indicator nuclide (n,γ) reaction rate of graphite specimens, which impact both plutonium production and the irradiation of indicator nuclides.

The FOM values for parameters were compared between the HANARO full core model with and without variance reduction. FOMs in the model with variance reduction consistently exceeded those without it for all parameters. FOM ratios, denoting the FOM from the model with variance reduction divided by that from the model without it, are described in Fig. 4 and Fig. 5. Fig. 4 presents the parameters outcomes for nuclear fuel specimens. The flux FOM ratio of nuclear fuel specimens ranged approximately between 80 and 110, while the FOM ratio for the ^{238}U (n,γ) reaction rate varied considerably but consistently exceeded 1, reaching up to 80. Fig. 5 specifically shows the ratios of parameters for the Rod2 graphite specimen. The FOM ratio for the Flux and Boron, Iron, and Titanium reaction rate of Rod2 graphite ranged between 20 and 50, while the remainder ranged between 10 and 30. The Rod3 results are not graphically represented, but the trend is similar to Rod2 data.

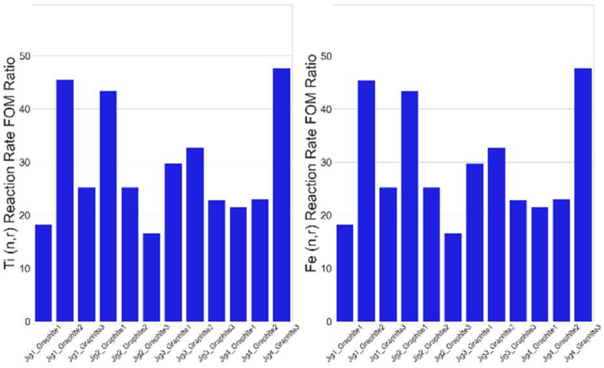


(a) flux *(b) ^{238}U*

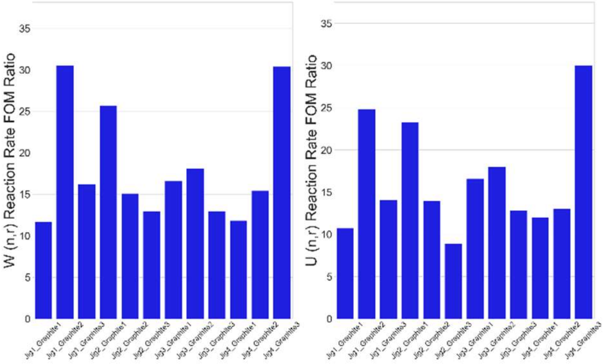
Fig. 4. FOM ratio comparison for each fuel nuclear specimen, (a) flux, (b) ^{238}U (n,γ) reaction rate FOM ratio graph



(a) flux *(b) Boron*



(c) Titanium *(d) Iron*



(e) Tungsten *(f) Uranium*

Fig. 5. FOM ratio comparison for each graphite specimen in rod2, (a) flux, (b) Boron, (c) Titanium, (d) Iron, (e) Tungsten, (f) Uranium (n,γ) reaction rate FOM ratio graph

Through these results, it can be determined that the model with variance reduction is suitable for uncertainty analysis. The cumulative plutonium data from 100 independent simulation results were sampled by Jig, while the graphite tally data was organized by Jig and individual rods.

2.4 Polynomial Regression

The polynomial regression method with a logarithmic transformation, using sampling data, is utilized to estimate the prediction line based on Jig and individual rods. The least squares method is implemented in Eq. (2).

$$f(x) = \sum_i^n a_i (log x)^i \qquad (2)$$

Function $f(x)$ represents the mass density of plutonium. x is the isotope ratio data of an indicator element obtained from McCARD. In this study, the regression order is set to 3rd.

Fig. 6 shows the prediction line and sampling data using B-10/B-11 for the case of the statistical factor. The blue dots represent cumulative plutonium sampling data, while the red line depicts the prediction line using 3rd polynomial regression with a logarithmic transformation. The graphs display sample data for 23 burnup points ranging from 28 to 336 Effective Full Power Days (EFPDs). Except for the first cycle, the data for 5 and 28 days were sampled in each cycle. Each subfigure displays plutonium data categorized by sampling locations such as Jigs and individual rods. The coefficients of determination (R^2) for the regression exceeded 0.997, regardless of indicator nuclides and sampling location. The detailed coefficients of determination are shown in Table 3. Fig 7 shows results using manufacturing tolerance factors in a format similar to that of Fig 6. Furthermore, the coefficients of determination exceeded 0.996, indicating the suitability of the polynomial regression function. The coefficients of determination are arranged in Table 4.

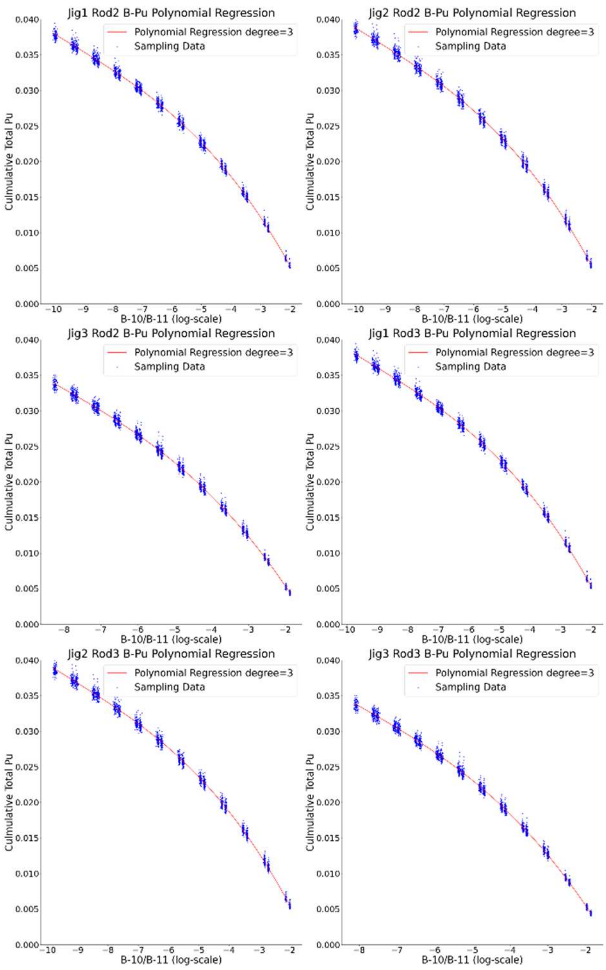


Fig. 6. Prediction line using 3rd polynomial regression and B-10/B-11 sampling data by stochastic influence.

Table. 3: Coefficients of determination summary table by stochastic factor.

Indicator nuclides	index	R^2 with Rod2	index	R^2 with Rod3
Boron	Jig1	0.99796	Jig 1	0.99801
	Jig 2	0.99754	Jig 2	0.99756
	Jig 3	0.99735	Jig 3	0.99733
Titanium	Jig1	0.99798	Jig 1	0.99803
	Jig 2	0.99757	Jig 2	0.99758
	Jig 3	0.99736	Jig 3	0.99735
Iron	Jig1	0.99798	Jig 1	0.99803
	Jig 2	0.99757	Jig 2	0.99758
	Jig 3	0.99736	Jig 3	0.99734
Tungsten	Jig1	0.99769	Jig 1	0.99801
	Jig 2	0.99755	Jig 2	0.99747
	Jig 3	0.99737	Jig 3	0.99736
Uranium	Jig1	0.99769	Jig 1	0.99801
	Jig 2	0.99754	Jig 2	0.99756
	Jig 3	0.99735	Jig 3	0.99733

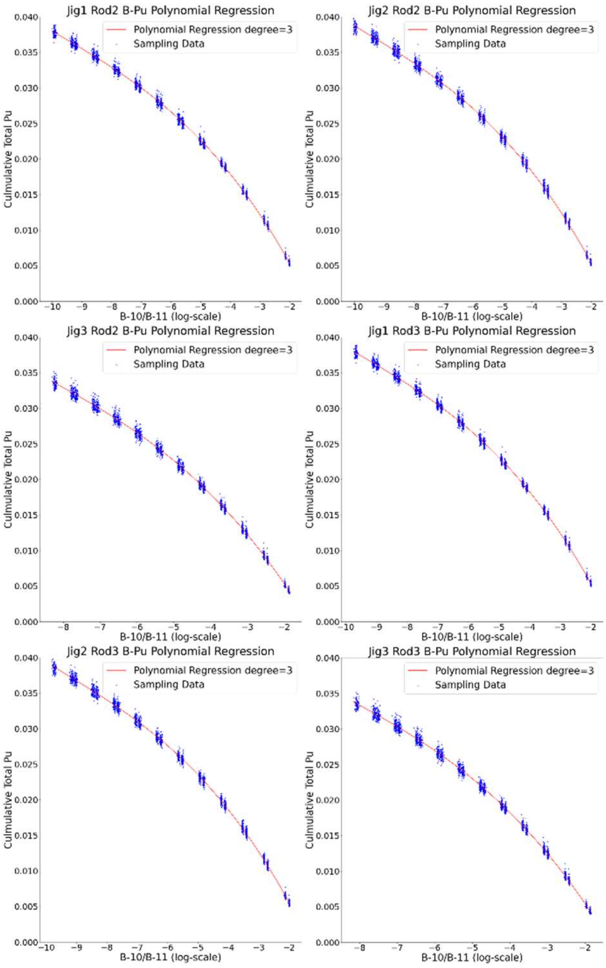


Fig. 7. Prediction line using 3rd polynomial regression and B-10/B-11 sampling data by stochastic and manufacturing tolerance influence.

Table. 4: Coefficients of determination summary table by stochastic and manufacturing tolerance table

Indicator nuclides	index	R^2 with Rod2	index	R^2 with Rod3
Boron	Jig1	0.99818	Jig 1	0.99819
	Jig 2	0.99781	Jig 2	0.99784
	Jig 3	0.99705	Jig 3	0.99701
Titanium	Jig1	0.99820	Jig 1	0.99821
	Jig 2	0.99784	Jig 2	0.99786
	Jig 3	0.99706	Jig 3	0.99701
Iron	Jig1	0.99820	Jig 1	0.99821
	Jig 2	0.99784	Jig 2	0.99786
	Jig 3	0.99706	Jig 3	0.99701

Tungsten	Jig1	0.99821	Jig 1	0.99821
	Jig 2	0.99775	Jig 2	0.99785
	Jig 3	0.99694	Jig 3	0.99705
Uranium	Jig1	0.99818	Jig 1	0.99819
	Jig 2	0.99781	Jig 2	0.99784
	Jig 3	0.99705	Jig 3	0.99700

3. Results

The residuals between predicted data and sampled data were used for uncertainty estimation. The relative error of residuals was analyzed according to burnup.

3.1 Stochastic error of MC code

Normalized root mean square error (NRMSe) was used to estimate the distribution of residuals and is shown in Eq (3)

$$NRMSe = \sqrt{\frac{1}{n-1}\frac{\sum_i^n (y_{p,i}-y_{e,i})}{\overline{y_e}^2}} \quad (3)$$

y_p is the predicted cumulative plutonium mass density and y_e is the sampled cumulative plutonium mass density obtained from McCARD. The NRMSe for each indicator element sampling data, ranging from 28 to 336 days, is presented in Fig. 8. The subfigures illustrate the estimation results according to sampling locations.

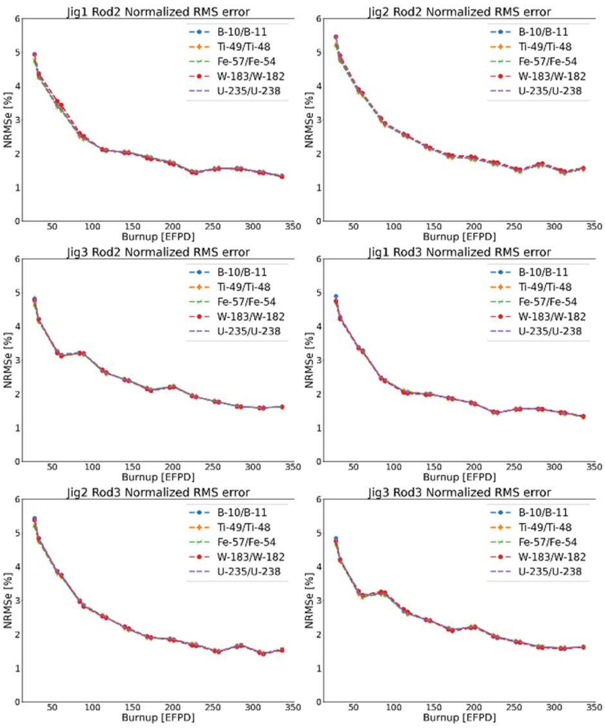


Fig. 8. Comparison of NRMSe of total indicator nuclides by stochastic error by operation

3.2 Manufacturing tolerance of experimental instrument

Especially, the total error of the manufacturing tolerance factor by sampling method includes the stochastic error of MC code because random sampling of the dimensions according to the manufacturing tolerance leads to a different random sequence in McCARD. The uncertainty estimation for the case of the manufacturing tolerance factor was followed in the same procedure as for the stochastic error. The NRMSe resulting from the stochastic error and manufacturing tolerance factors is shown in Fig. 9.

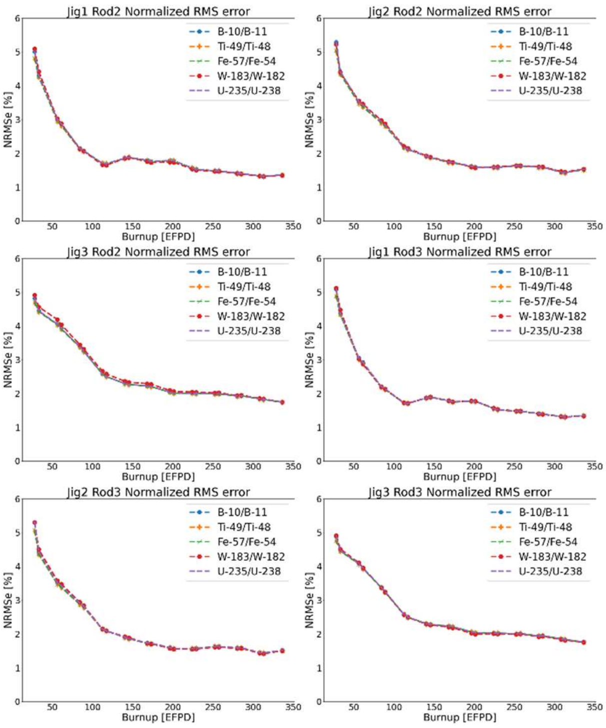


Fig. 9. Comparison of NRMSe of total indicator nuclides by stochastic error and manufacturing tolerance by operation

4. Conclusions

In this study, the HANARO neutron irradiation experiments were simulated using the precise HANARO model. A simulation model was developed using the variance reduction method and the sampling data obtained from depletion calculation using McCARD were compared to the predicted data derived from a polynomial regression function with a logarithmic transformation. The NRMSe by statistical factor ranged from approximately 5 to 6 % at 28 days and decreased to about 1% by 336 days independently of indicator elements. Furthermore, The NRMSe resulting from the stochastic error of MC code and manufacturing tolerance factors were comparable to that of the stochastic error alone. It was estimated that the influence of the stochastic error factor is more significant than that of the manufacturing tolerance factor. Therefore, it was concluded that the influence of the manufacturing tolerance factor is negligible, and total uncertainty could be described as shown below in Eq (4). σ_r is uncertainty stochastic error and σ_r is uncertainty by manufacturing tolerance.

$$\sigma^2 = {\sigma_r}^2 + {\sigma_t}^2 \approx {\sigma_r}^2 \quad (4)$$

Furthermore, it is planned to evaluate how the covariance of nuclear cross-sectional data is going to impact the final experimental uncertainty.

Acknowledgments

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

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