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Establishment of The Evaluation Criteria For Swipe Environmental Samples From Nuclear Facilities

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ABSTRACT

The International Atomic Energy Agency (IAEA) monitors undeclared nuclear activities in member states by environmental sampling program. The Republic of Korea (ROK), as a member state of the IAEA, has also been operating a domestic environmental sampling program. The environmental sampling program in the ROK includes the collection, screening, and particle analysis of cotton swipe samples. However, there are no evaluation criteria for the analysis results to verify undeclared nuclear activities. Therefore, we intended to establish evaluation criteria using a reference spent fuel rod and the MCNP6 and Origen-Arp code. The reference spent fuel rod has the following characteristics: PLUS7 type assembly (CE 16x16), 4.5 wt.% enrichment, 45 GWD/MTU discharge burnup, and 30 years cooled after discharge. The spent nuclear fuel was divided into 52 sections in the axial direction to obtain the average fission energy deposition of neutron for each section using MCNP6. The discharged burnup for each section was estimated based on the fission energy deposition of neutrons. The isotopic ratio ($^{234}U/^{238}U$, $^{241}Pu/^{240}Pu$, $^{234}U/^{236}U$, $^{238}Pu/^{239}Pu$, and $^{236}Cm/^{244}Cm$) of each section was calculated using Origen-Arp. We then calculated the range of the isotopic ratio using the simulation results. Thereafter, we calculated the isotopic ratio of controlled fuel rods using different enrichment, discharge burnup, and cooling time. The isotopic ratios between the reference rod and controlled rods were compared using the t-test. As a result, we have established the evaluation criteria of swipe samples from a spent fuel assembly and will use them as irradiation information is given.

INTRODUCTION

The International Atomic Energy Agency (IAEA) recommended the environmental sampling program as an effective method to enhance nuclear safety through the '93+2 program' [1]. Environmental sampling is based on the assumption that even trace amounts of nuclides are released from nuclear facilities. Cotton swipe samples on the surface of equipment and facilities or external soil and water are collected to trace nuclear activities. The IAEA monitors undeclared nuclear activities in member states by environmental sampling. For example, samples are collected from a facility that handles uranium that is less than 5% enriched and these samples are then analyzed to assess if more than 5% enriched uranium is detected. South Korea, as a member state of the IAEA, has also been operating a domestic environmental sampling program. We sought to establish evaluation criteria for verifying the analysis results since there are no criteria in Korea. For this purpose, computing simulation programs (MCNP6 and Origen-ARP) were used to simulate spent nuclear fuel under specific conditions [2, 3]. Statistical methods were applied to the calculated isotope ratio to evaluate whether it could be used as an evaluation factor according to each condition. The isotopes used in this paper are those that can be analyzed using a large geometry secondary ion mass spectrometer (LG-SIMS) and whose correlation coefficient |r| between factors (burnup, cooling time, and enrichment of fresh fuel) calculated using Origen is higher than 0.7.

EXPERIMENTAL PROCEDURES

Figure 1 shows a simplified structure of the active fuel part in a PLUS7 fuel rod. This geometry was entered into MCNP6 and divided into 52 sections in the axial direction. The reference spent fuel rod has the following conditions: 4.5 wt.% enrichment, 45 GWD/MTU discharge burnup, 3 x 12 months of fuel cycle with 2 x 60 days of interval overhaul, and 30 years passed after discharge. F7 tally (average fission energy deposition of neutron) was used to estimate the discharge burnup of each section. The number of sections was increased from 52 to 200 by interpolation and the five highest and lowest values were removed to match 95 % confidence (k=2). The estimated discharge burnup for 20 out of 190 sections was then entered into Origen-Arp to obtain the isotope ratios of $^{234}U/^{238}U$, $^{241}Pu/^{240}Pu$, $^{234}U/^{236}U$, $^{238}Pu/^{239}Pu$, and $^{236}Cm/^{244}Cm$, which were suggested as indicators for spent fuel irradiation characteristics in a previous study [4]. The isotope ratios according to the enrichment (4.0 and 5.0 wt.%), discharge burnup (35, 50, and 55 GWD/MTU), and cooling time (25 and 35 y) were compared with the reference spent fuel. An F-test and t-test were performed to evaluate whether the isotope ratio could be used as an evaluation factor according to each condition [5]. The F-test is used to determine the difference between the standard deviations of two groups. The F value is calculated as

$$F=s_1^2/s_2^2 \ (s_1>s_2) \tag{1}$$

where s is the standard deviation. If the calculated F-value is greater than the critical value (F_{table}) when two groups have 95 % confidence (i.e. 2.168), then it indicates that the probability that the two groups were obtained from a population with the same standard deviation is less than 5 %. It was found that standard deviations of the two groups differed significantly. The t-test meanwhile is a method to determine significance by comparing the mean values of two groups. The formula for calculating t depends on the results of the F-test. If the F value is less than the F_{table}, t is calculated as

$$s_p = \sqrt{\frac{s_1^2\times(n_1-1)+\ s_2^2\times(n_2-1)}{n_1+\ n_2-2}} \tag{2}$$

$$t = \frac{|\overline{x_1}-\overline{x_2}|}{s_p}\sqrt{\frac{n_1\times n_2}{n_1+n_2}} \tag{3}$$

where n is the number of data in a group and x is the mean value. In the opposite case ($F > F_{table}$), t and D_f are calculated as follows:

$$t = \frac{|\overline{x_1} - \overline{x_2}|}{\sqrt{(s_1^2/n_1) + (s_2^2/n_2)}} \tag{4}$$

$$D_f = \frac{(s_1^2/n_1 + s_2^2/n_2)^2}{\frac{(s_1^2/n_1)^2}{n_1 - 1} + \frac{(s_2^2/n_2)^2}{n_2 - 1}} \tag{5}$$

If the $t_{calculated}$ is higher than the critical value (t_{table}), it means that the two groups have significance, and this isotope ratio then can be used as an evaluation factor.

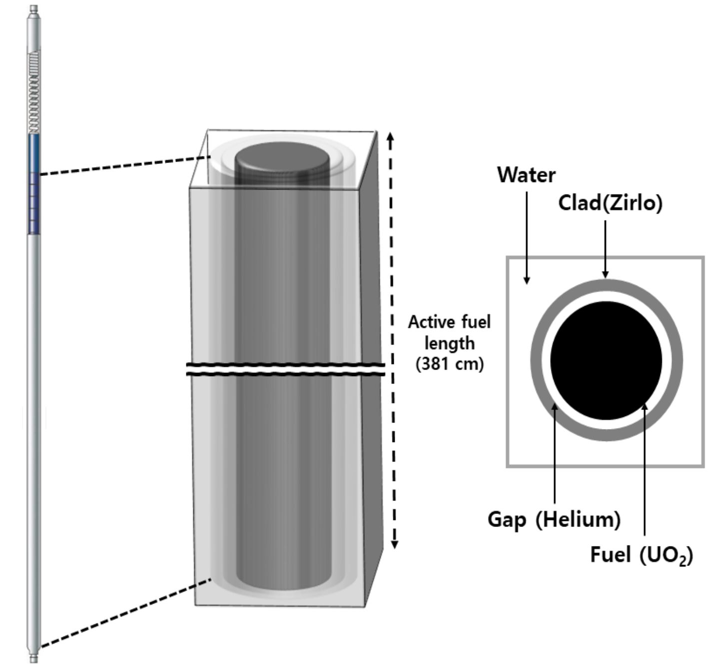

Figure 1. Schematic drawing (left) and cross-section (right) of a PLUS7 fuel rod

RESULTS & DISCUSSION

Figures 2 and 3 show the estimated discharge burnup of each section in a reference spent fuel rod and the isotope ratio when 30 y have passed after discharge, respectively. An F-test and t-test were performed on the reference conditions and the results of changing burnup, cooling time, and enrichment. In each case, the conditions other than that to be changed were the same as the reference conditions.

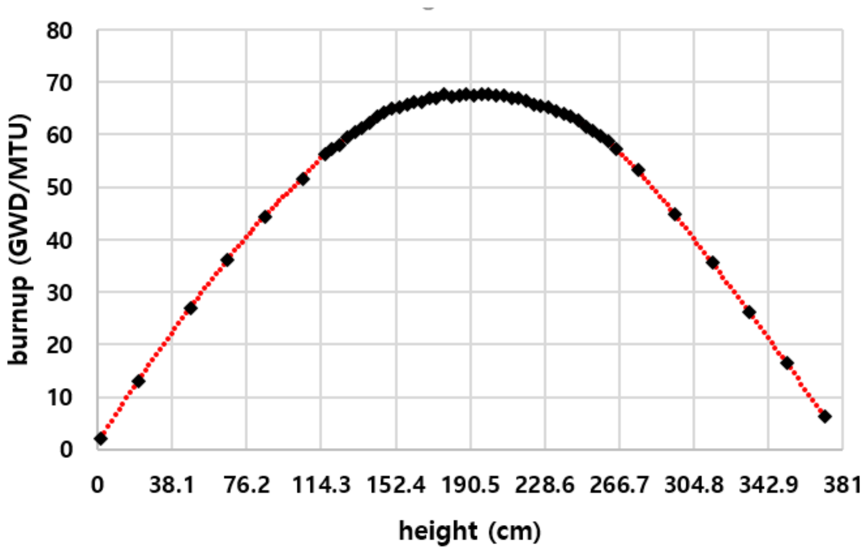

Figure 2. Calculated (diamond) and interpolated (dot) discharge burnup of reference spent fuel rod

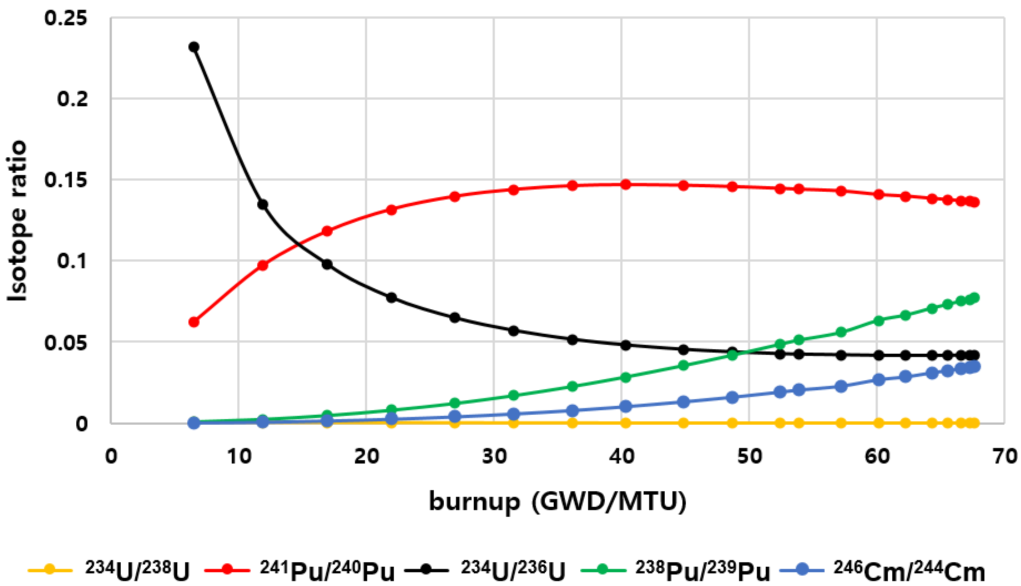

Figure 3. Isotope ratio of actinides when 30 y passed after discharge

Burnup

The F-test and t-test results of the average burnup of 35, 50, and 55 GWD/MTU, as well as those of the reference condition, 45 GWD/MTU, are listed in Table 1. According to Table 1 (a), none of the isotope ratios show significance in the average burnup of 5 GWD/MTU. There was a significance in the ratio of ^{238}Pu/^{239}Pu from the 10 GWD/MTU difference in average burnup (Tables 1 (b) and (c)). Therefore, there is a possibility that ^{238}Pu/^{239}Pu can be used as an evaluation factor to determine the average burnup condition of spent fuel.

Table 1. The F-test and t-test results of the average burnup of (a) 50, (b) 55, and (c) 35 GWD/MTU, and the reference condition, 45 GWD/MTU

(a)	$^{234}U/^{238}U$	$^{241}Pu/^{240}Pu$	$^{234}U/^{236}U$	$^{238}Pu/^{239}Pu$	$^{246}Cm/^{244}Cm$
F	1.183	1.274	1.289	1.466	1.661
>F_{table}	O	O	O	O	O
s_p	2.807E-05	2.183E-02	4.958E-02	2.545E-02	1.138E-02
t	0.729	0.049	0.972	1.772	1.885

(b)	$^{234}U/^{238}U$	$^{241}Pu/^{240}Pu$	$^{234}U/^{236}U$	$^{238}Pu/^{239}Pu$	$^{246}Cm/^{244}Cm$
F	1.437	1.501	1.585	1.817	2.487
>F_{table}	O	O	O	O	X
s_p	2.474E-05	1.869E-02	4.184E-02	3.293E-02	-
D_f	38 (t_{38} = 2.024)				33.6 (t_{33} = 2.035)
t	0.466	1.134	1.159	**2.268**	1.928

(c)	$^{234}U/^{238}U$	$^{241}Pu/^{240}Pu$	$^{234}U/^{236}U$	$^{238}Pu/^{239}Pu$	$^{246}Cm/^{244}Cm$
F	1.342	1.605	1.681	2.514	3.283
>F_{table}	O	O	O	X	X
S_p	2.474E-05	1.869E-02	4.184E-02	-	-
D_f	38 (t_{38} = 2.024)			32.271 (t_{32} = 2.037)	29.717 (t_{29} = 2.045)
t	1.278	0.154	0.660	**2.174**	**2.385**

Cooling time

Tests results for cooling time of 25 and 35 y, and the reference condition, 30 y, are listed in Table 2. There was a significance in the ratio of $^{241}Pu/^{240}Pu$ from the 5 y difference in cooling time after discharge. Therefore, the ratio of $^{241}Pu/^{240}Pu$ can be used as an evaluation factor for cooling time.

Table 2. The F-test and t-test results of the average burnup of (a) 25 and (b) 35 y, and the reference condition, 30 y

(a)	$^{234}U/^{238}U$	$^{241}Pu/^{240}Pu$	$^{234}U/^{236}U$	$^{238}Pu/^{239}Pu$	$^{246}Cm/^{244}Cm$
F	1.499	1.637	1.031	1.082	1.464
>F_{table}	O	O	O	O	O

s_p	3.004E-05	2.351E-02	4.670E-02	2.831E-02	1.166E-02
D_f	38 (t_{38} = 2.024)				
t	1.930	3.964	0.589	0.767	1.605

(b)	$^{234}U/^{238}U$	$^{241}Pu/^{240}Pu$	$^{234}U/^{236}U$	$^{238}Pu/^{239}Pu$	$^{246}Cm/^{244}Cm$
F	1.477	1.634	1.029	1.082	1.464
>F_{table}	O	O	O	O	O
s_p	2.460E-05	1.838E-02	4.601E-02	2.721E-02	1.411E-02
D_f	38 (t_{38} = 2.024)				
t	2.089	3.965	0.584	0.769	1.605

Enrichment

Tests results of average burnup of 4.0 and 5.0 wt.%, and the reference condition, 4.5 wt.%, are listed in Table 3. There was a significance in the ratio of $^{234}U/^{238}U$ from the 0.5 wt.% difference in enrichment of fresh fuel. Therefore, the ratio of $^{234}U/^{238}U$ can be used as an evaluation factor for enrichment.

Table 3. The F-test and t-test results of the average burnup of (a) 4.0 and (b) 5.0 wt.%, and the reference condition, 4.5 wt.%

(a)	$^{234}U/^{238}U$	$^{241}Pu/^{240}Pu$	$^{234}U/^{236}U$	$^{238}Pu/^{239}Pu$	$^{246}Cm/^{244}Cm$
F	1.565	1.349	1.420	1.050	1.151
>F_{table}	O	O	O	O	O
s_p	2.433E-05	1.910E-02	4.277E-02	2.741E-02	1.318E-02
D_f	38 (t_{38} = 2.024)				
t	3.861	1.248	0.941	0.677	1.252

(b)	$^{234}U/^{238}U$	$^{241}Pu/^{240}Pu$	$^{234}U/^{236}U$	$^{238}Pu/^{239}Pu$	$^{246}Cm/^{244}Cm$
F	1.382	1.187	1.168	1.126	1.359
>F_{table}	O	O	O	O	O
s_p	2.933E-05	2.141E-02	4.825E-02	2.696E-02	1.184E-02
D_f	38 (t_{38} = 2.024)				
t	3.486	1.387	0.622	0.869	1.363

CONCLUSIONS

Evaluation criteria for swipe samples from nuclear facility were established using MCNP6 and Origen-Arp. The reference spent fuel rod used for the simulation has the following characteristics: a PLUS7 type assembly (CE 16x16), 4.5 wt.% enrichment, 45 GWD/MTU discharge burnup, and 30 years passed after discharge. The spent nuclear fuel was divided into 52 sections in the axial direction to obtain the average fission energy deposition of neutrons for each section using MCNP6. The discharge burnup was calculated based on the fission energy deposition of neutrons. The isotopic ratio of each section over cooling time was calculated using Origen-Arp. The evaluation factors were then determined by an F-test and t-test between the reference conditions and a control group. As a result, the ratios of $^{238}Pu/^{239}Pu$, $^{241}Pu/^{240}Pu$, and $^{234}U/^{238}U$ can be used as evaluation factors for burnup, cooling time, and enrichment, respectively. We can use these factors as irradiation information is provided.

REFERENCES

[1] IAEA, Model Protocol Additional to the Agreement(s) between State(s) and the International Atomic Agency for the Application of Safeguards, INFCIRC/540 corr., 1997.

[2] Christopher J. Werner et al., MCNP® User’s Manual Code Version 6.2, Los Alamos National Laboratory, LA-UR-17-29981, February 2018.

[3] S. M. Bowman and I. C. Gauld, OrigenArp Primer: How to Perform Isotopic Depletion and Decay Calculations with SCALE/ORIGEN, ORNL/TM-2010/43, April 2010.

[4] Haneol Lee and Hwan Lee, Identification of Parameters for Evaluating Swipe Samples in Nuclear Safeguards, 4th Conference on Nuclear Analytical Techniques & 6th Symposium in Medicine, Space and Power, Nov, 12-13, 2020.

[5] Danial C. Harris, Quantitative Chemical Analysis 9th edition, W. H. Freeman, NY, 2015.