Uncertainty in determination of the Pu-to-^{244}Cm-ratio in pyroprocess

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1. Introduction

According to the IAEA safeguards glossary report, the objective of safeguards is that *"the timely detection of diversion of significant quantities of nuclear material from peaceful nuclear activities to the manufacture of nuclear weapons or of other nuclear explosive devices or for purposes unknown, and deterrence of such diversion by the risk of early detection"* [1]. By that, it is considered that material accountancy in nuclear facilities should be established for safeguards.

One of strategies for the Pu material accounting in pyroprocess is the Pu-to-^{244}Cm-ratio method [2] which is based on the below equation,

$$\left(\text{Pu mass}\right)=\left(\frac{\text{Pu}}{^{244}\text{Cm}}\right)\times\left(^{244}\text{Cm mass}\right). \quad (1)$$

The ratio of Pu-to-^{244}Cm is determined by DA (destructive assay), NDA (non-destructive assay), or numerical depletion simulation codes prior to the key-pyroprocess, which mainly consists of electrolytic-reduction, electro-refining, and electro-winning, etc. However, the inaccuracy of this method, particularly in determination of the ratio, could be caused by heterogeneity, non-uniformity, and error of instruments and codes, etc. In this study, how the non-uniformity of nuclide composition in spent fuel affects on the material accountancy is discussed.

2. Methods and Results

2.1 Characterization of representative spent fuel

Because there are 12 OPR(Optimized Power Reactor)s-1000 among 21 PWR(Pressurized Water Reactor)s in South Korea [3], 3 different types of nuclear assemblies are loaded in the OPR-1000 core. The difference among 3 types assemblies (Type-0, Type-1, and Type-2) is a number of GBF (Gadolinia-Bearing Fuel) and its loading location. In this study, the Type-0 fuel assembly is firstly selected as shown in Fig. 1. The detailed design parameters for that shown in the reference [4].

The depletion simulations for the representative nuclear fuel assemblies have been conducted by the SERPENT code [5]. In order to obtain the non-uniform composition in fuels in the assembly, the 30 axial meshes, which is limited by the CPU memory, for each fuel rod are discretely given. The moderator temperature variation in the axial direction is applied. The upper plenum in the fuel rod is modeled. It is assumed that 1) no boron in the coolant, 2) the 90 % capacity factor in 18 months for one cycle, 3) a constant power during the cycle, and 4) the 5-year cooling time after discharging.

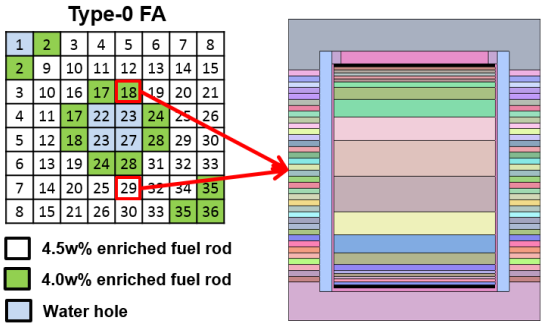

Fig. 1. Quarter of Type-0 nuclear fuel assemblies in OPR-1000 and its fuel pin-cell modeling by SERPENT.

In addition to achieve the radial non-uniformity in each fuel, the radial distribution for a fuel rod is predetermined by one more simulation. The lattice name for each fuel pin-cell is given with an Arabic number. Since the SERPENT system recognizes all equally enriched fuel pin-cells as one fuel pin-cell by giving identical lattice name for those, more meshes for the active fuel rod region can be given. By the simulation with this modeling, the representative radial nuclide distribution for each enriched fuel rod depending on the axial location can be obtained. The normalized radial distributions with respect the axial location are applied proportionally to the results of simulations shown in Fig. 1 by matching the results with the axial height of the fuel rods. In this way, the axial and radial non-uniformity of fuel rods in the assembly can be approximately achieved.

2.2 Statistical analysis to evaluate the uncertainty in determination of the Pu-to-^{244}Cm ratio

The head-end process consists of chopping, decladding voloxidation, and the granulation process [6]. Among those processes, the sampling to evaluate the ratio of Pu-to-^{244}Cm would be conducted after the granulation process. Therefore, the uncertainty in determination of ratio is evaluated by sampling of a

granule. The random variable, the ratio, consists of two random variables, the concentrations of Pu and ^{244}Cm as shown below

$$RV_{Ratio} = \frac{RV_{Pu}}{RV_{^{244}Cm}} \quad . \tag{2}$$

First, let us evaluate means and standard deviations of Pu and ^{244}Cm concentrations in a granule and its distribution. The voloxidation process produces powders in the micrometer scale, then multiple powders become one granule in the centimeter scale by the deoxidation in the pretreatment. It is not possible to model the mesh size for the active fuel region equal to the size of powder, therefore, it is assumed that the nuclide composition is uniformly distributed within each mesh. The sizes of each powder and granule are respectively identical. In this case, a number of powders to be one granule is large. By that, the standard deviation of nuclide composition in the single granule could be evaluated by the central limit theorem based on a law of large number as shown in Eq. 3, if the sizes of granule and powder are known. Then, we can define the two random variables, the concentration of Pu (RV_{Pu}) and ^{244}Cm ($RV_{^{244}Cm}$), following the normal distribution. The diameters of powder and granule are assumed 100 μm and 0.2 cm in the sphere shape so that the number of powders in the single granule is 8000 which is the denominator in the below equation,

$$\sigma_{Pu\text{ or }^{244}Cm} = \frac{\sigma_{Pu\text{ or }^{244}Cm\text{ in total powders}}}{\sqrt{\text{Number of powders in a granule}}} \quad . \tag{3}$$

Even though the distribution of two random variables, Pu and ^{244}Cm concentrations, are evaluated, there is one difficulty in the statistical analysis for the ratio of those two random variables. The reason is that the distribution for ratio of two normal distributions follows the Cauchy distribution for which the mean and the standard deviation are not calculable. Therefore, in this study, the normal approximation method for the ratio has been applied [7]. When the two random variables independent, the formula for the *Geary-Hinkley Transformation* is

$$RV_{Ratio}^{\ 2}\left(\mu_{^{244}Cm}^{\ 2} - z^2\sigma_{^{244}Cm}^{\ 2}\right) - 2RV_{Ratio}\left(\mu_{^{244}Cm}\mu_{Pu}\right) + \left(\mu_{Pu}^{\ 2} - z^2\sigma_{Pu}^{\ 2}\right) = 0 \,, \tag{4}$$

where z is 1.96 (95% confidence interval).

2.3 Results

The axial non-uniformity for Pu (top) and ^{244}Cm (bottom) mass density of each fuel rod has been plotted as a function of axial location (x-axis) depending on the different depletion period, 1 (blue lines), 2 (black lines), and 3 (red lines) cycle depletion periods as shown in Fig. 2. The significant axial non-uniformity is observed for all cases, particularly, at the top and bottom of the fuel rods. The axial non-uniformity of ^{244}Cm is more significant than that of Pu. Moreover, the increasing burnup could decrease the significance of axial non-uniformity.

By taking into account the radial non-uniformity and the voloxidation process effect, the statistical summary, the mean (μ) and the standard deviation (σ), for powders has been shown in Table. I.

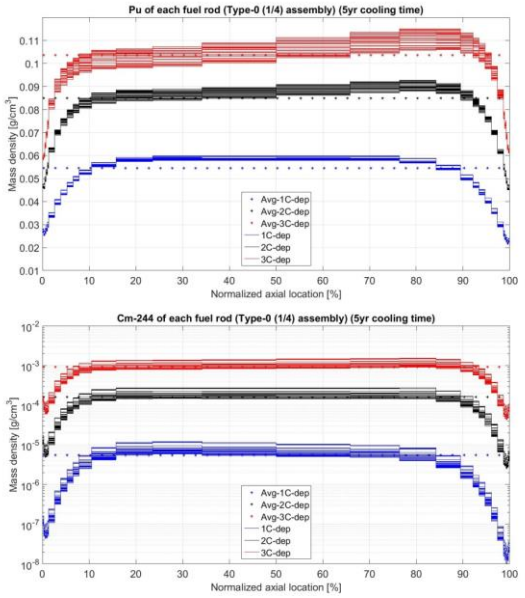

Fig. 2. Axial non-uniformity for Pu (top) and ^{244}Cm (bottom) as a function of axial location (x-axis) for 3 different depletion periods (blue: 1 cycle, black: 2 cycles, red: 3 cycles depletion).

Table I: Mean and standard deviation of Pu and ^{244}Cm mass density for powders after voloxidation

Cycle	Pu [g/cm^3]		^{244}Cm [g/cm^3]	
	μ	σ	μ	σ
1	5.45E-2	2.05E-2	5.52E-6	4.13E-6
2	8.49E-2	2.88E-2	1.60E-4	1.04E-4
3	1.04E-1	3.20E-2	9.28E-4	5.58E-4

Table II: Mean and standard deviation of Pu and ^{244}Cm mass density in the sampled single granule.

Cycle	Pu [g/cm^3]		^{244}Cm [g/cm^3]	
	μ	σ	μ	σ
1	5.45E-2	2.30E-4	5.52E-6	4.62E-8
2	8.49E-2	3.22E-4	1.60E-4	1.16E-6
3	1.04E-1	3.58E-4	9.28E-4	6.24E-6

The standard deviations in Table. I. are substituted into Eq. (3) to evaluate that of the single granule after the granulation process. The results are shown in Table. II. Because of the sample size which is a number of powders to be one granule, is large as 8000, the

standard deviation of Pu and ^{244}Cm mass densities in the single sample granule is dramatically decreased.

In order to evaluate the uncertainty of the ratio, the data in Table. II are substituted into Eq. 4. By solving quadratic equations with respect to RV_{Ratio} for each case, two values (RV_{Ratio_1} and RV_{Ratio_2}) are respectively calculated. Then, the point estimators for the mean (μ_{Ratio}) and the standard deviation (σ_{Ratio}) of ratio are evaluated by below,

$$\mu_{Ratio} = \left(RV_{Ratio\_1} + RV_{Ratio\_2}\right)/2, \qquad \sigma_{Ratio} = \left|RV_{Ratio\_1} - RV_{Ratio\_2}\right|/z. \tag{5}$$

The summary of results is shown in Table. III. First, the ratio is significantly varied depending on the depletion length. The coefficient of variances (CV), which is evaluated by the standard deviation to the mean, range between 1 to 2 %, when the sizes of powders and granules are fixed as 100 μm and 0.2 cm. However, it could decrease by taking samples that shows i) the size of granule is greater than 0.2 cm, and ii) the size of powder is smaller than 100 μm in that granule.

Table. III: Uncertainty in determination of the ratio evaluated using the *Geary-Hinkley Transformation* method.

Cycle	Pu-to-^{244}Cm-ratio		
	μ_{Ratio}	σ_{Ratio}	CV [%]
1	9.88E+3	1.85E+2	1.87
2	5.32E+2	8.73E+0	1.64
3	1.12E+2	1.69E+0	1.51

3. Conclusions

The methodology to analysis how the non-uniformity of nuclide composition in spent fuel affects on material accountancy, while the Pu-to-^{244}Cm-ratio method is being applied. In this study, the result of uncertainty in determination of the ratio is conducted based on the limited assumptions. Therefore, we need to analysis more various cases which would be able to cover the practical processing situation to discuss on the material accountancy for pyroprocess, for instance, other types of spent fuel assemblies and the wide spectrum of sizes of powder and granules.

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