Input Accountancy of Pyroprocessing With Different Head-end Process Options

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1. Introduction

In the prospective of spent nuclear fuel management, pyroprocessing is one of the options to reduce the volume and toxicity of spent nuclear fuel waste [1]. To secure proliferation resistance in spent nuclear fuel recycling, safeguards approaches should be developed. Among material accountancy, input material in the head-end process of pyroprocessing, which is the Material Balance Area (MBA) to declare Shipper-Receiver Difference (SRD), is an issue because materials exist in the solid form such as small and large fragment, powder, and/or porous pellet which can increase the sampling uncertainty when taking Destructive Analysis (DA) samples. Material forms and size largely depend on the head-end process. Regardless of the head-end process, input material accounting methods should be established. In this study, two input accounting methods according to the head-end process options such as decladding methods, spent nuclear fuel particle size, and process equipment were proposed and evaluated.

2. Double Stage Homogenization

In the previous work [2], double stage homogenization was proposed in case that the material form is powder and the performance of homogenization mixers with metal oxide powder as surrogate material was evaluated in terms of heterogeneity, sampling uncertainty, and accounting uncertainty. The tumbler mixer known as a diffusive-type mixer may have poor heterogeneity in case of mixing powder with different sizes and/or different density. In this study, to check the segregation effect, the tumbler mixer of 50 kg capacity was tested with depleted uranium powder and metal oxide power of various particle sizes as shown in figure 1. DA samples were taken in 10 different positions of the tumbler mixer container at each mixing time, and analyzed by a titration method to measure the mass of total uranium in the samples. The analyzed heterogeneity was less than 0.5% which was similar to the previous test in which the metal oxide powder of the same particle size was mixed. It means that the heterogeneity of the diffusive mixer such as tumbler mixers is barely affected by segregation with powder of 10 to 75 um particle size.

Fig. 1. Depleted uranium powder of 10 um & 40 um tumbler mixer.

3. Representative sampling & Homogenization

In input accountancy with the double homogenization process, it was assumed that the oxidative decladding is used and whole spent nuclear fuel is pulverized by the voloxidation process. Thus, the sampling uncertainty can be reduced by homogenization mixing of spent nuclear fuel powder. On the other hand, in case of using the mechanical decladding instead of the oxidative decladding, the decladded spent nuclear fuel exists as small and large fragments which cannot be homogenized by mixing. Thus, combination of representative sampling and homogenization was proposed for input accountancy. The description of the head-end process is shown in figure 2. Spent nuclear fuel fragment were sieved to separate small and big fragments, and a sample up to

30 kg from big fragments is taken. Then, the sample is pulverized by the voloxidation process, then final samples of 1 g are taken for DA analysis after homogenization mixing. Small fragments and spent nuclear fuel remained in hulls go into the voloxidation process followed by the homogenization process, and DA samples taken same as sampling of big fragment.

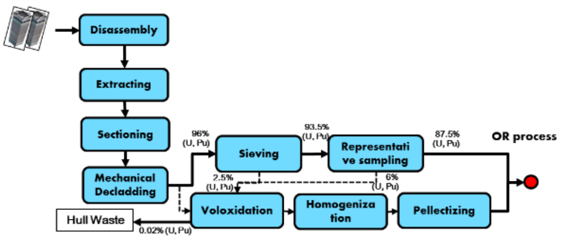

Fig. 2. Diagram of the head-end process.

In addition to the uncertainty arising from the homogenization process such as the heterogeneity, DA uncertainty, and the distribution of Pu concentration in spent nuclear fuel, a combination of the representative sampling and homogenization causes the sampling uncertainty which largely depends on the particle size, the sampling mass, and the performance of the sampling device. The performance of the sampling device, called the sample divider, was evaluated in previous work [3].The sampling uncertainty was similar to the theoretical value (binomial distribution) which is a function of particle size/density, and the number of samples to be divided, as the below equation [4, 5]. M is the sample mass, f_i is the overall mass fraction of component i, ω_i is the mass of a single particle of the component, and $\bar{\omega}$ is the overall mean particle mass in the equation.

$$(\sigma_m^2)_i = \frac{f_i(1-f_i)\omega_i + f_i^2(\bar{\omega}-\omega_i)}{M} \quad (1)$$

By the mechanical decldding of PWR spent nuclear fuel, the particle size is roughly smaller than 2 mm, thus the sample mass should be at least 34 kg to achieve the sampling uncertainty of 1 % with 99.7 % confidence. The sampling uncertainty with respect to the particle size will be more discussed in detailed in the conference after the statistical model calculation.

4. Discussion

Double stage homogenization and a combination of homogenization and representative sampling were proposed as Input accounting methods, depending on the head-end process options. The candidate mixer has no meaningful segregation effect to degrade the heterogeneity. Thus, The Pu accounting uncertainty of 1% (1σ) with the double stage homogenization can be achievable as estimated in the previous work [4]. And in case of taking enough sample mass from the fragment with the representative sampling device, the representative sampling can achieve a low sampling uncertainty which depends on the sampling mass, and particle size. Regardless of the head-end process, it is expected that nuclear material accounting in the head-end process with a low uncertainty is possible with the proposed methods.

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