



Original Article

Evaluation of nuclear material accountability by the probability of detection for loss of Pu (LOPu) scenarios in pyroprocessing

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ABSTRACT

A new methodology to analyze the nuclear material accountability for pyroprocessing system is developed. The Pu-to-²⁴⁴Cm ratio quantification is one of the methods for Pu accountability in pyroprocessing. However, an uncertainty in the Pu-to-²⁴⁴Cm ratio due to the non-uniform composition in used fuel assemblies can affect the accountability of Pu. A random variable, LOPu, is developed to analyze the probability of detection for Pu diversion of hypothetical scenarios at a pyroprocessing facility considering the uncertainty in Pu-to-²⁴⁴Cm ratio estimation. The analysis is carried out by the hypothesis testing and the event tree method. The probability of detection for diversion of 8 kg Pu is found to be less than 95% if a large size granule consisting of small size particles gets sampled for measurements. To increase the probability of detection more than 95%, first, a new Material Balance Area (MBA) structure consisting of more number of Key Measurement Points (KMPs) is designed. This multiple KMP-measurement for the MBA shows the probability of detection for 8 kg Pu diversion is greater than 96%. Increasing the granule sample number from one to ten also shows the probability of detection is greater than 95% in the most ranges for granule and powder sizes.

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1. Introduction

Nuclear material accountability (NMA) is a fundamental nuclear safeguards measure instituted by the International Atomic Energy Agency (IAEA) for nuclear facilities [1]. The material unaccounted for (MUF) is a characteristic that needs to be quantified for any NMA [1–6]. The MUF can be mathematically expressed as

$$MUF = (PB + X - Y) - PE \quad (1)$$

where *PB* and *PE* are the physical inventory at the beginning and ending of a given Material Balance Period (MBP), *X* and *Y* are respectively the sum of increases to inventory and decreases from inventory in a designated Material Balance Area (MBA) of a facility for the respective MBP [1]. Since the terms in Eq. (1) are the measured values, the MUF can be declared as a random variable and can be represented by statistical parameters, for instance, the mean (μ) and variance (σ^2). The formulation to evaluate the

standard deviation of MUF has been developed to discuss material accountability for a pyroprocessing system [5,6]. Fundamentally, in those references, the recommended plutonium (Pu) material accounting method is the Pu-to-²⁴⁴Cm ratio method [7] based on,

$$Pu_{KMP} = \left(\frac{Pu}{^{244}Cm} \right) \times ^{244}Cm_{KMP} \quad (2)$$

where Pu_{KMP} and $^{244}Cm_{KMP}$ are the respective masses of Pu and curium-244 (²⁴⁴Cm) at a key measurement point (KMP). The Pu-to-²⁴⁴Cm ratio assumed to remain constant through various steps of pyroprocessing such as the electrolytic-reduction, electro-refining, and electro-winning processes [5,8]. There is a possibility for this ratio not to remain constant in the electro-refining step of pyroprocessing due to minor electrical potential difference between Pu and Cm. This aspect has been discussed by Gonzalez et al. using a numerical approach [9]. Gonzalez et al. paper also mentioned the necessity of experimental validation to ascertain that the ratio need not remain a constant. However, in this study, the Pu-to-²⁴⁴Cm is assumed to be a constant during pyroprocessing. This ratio can be measured by measuring Pu and ²⁴⁴Cm masses in a sample [10]. In this study, it is assumed sampling is conducted

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at the granulation step of pyroprocessing. The reasons are follows: 1) the nuclide composition in granules samples could be more uniform than that in the chopped piece samples and 2) the sampling granules (a centimeter- or millimeter-scale) would be easier than the sampling of powders after voloxidation because the size of powders is in a micro-scale. At the final step of pyroprocessing where a transuranic-uranium (TRU:U) ingot is obtained as the product only $^{244}\text{Cm}_{\text{KMP}}$ mass is measured using a coincidence neutron detection system [11–15]. Even though ^{244}Cm can vary as functions of burnup, cooling time, type of a nuclear fuel and reactor type, the most dominant neutron source in used nuclear fuel assemblies within a cooling time of 3–5 years for reprocessing is ^{244}Cm . For instance, about 97% neutrons in a used fuel assembly discharged by a light water reactor with 45GWd/MTU are generated by a spontaneous fission of ^{244}Cm [7]. Therefore, the neutron counting and ^{244}Cm mass measurement can be applied for safeguarding the pyroprocessing system. With the $^{244}\text{Cm}_{\text{KMP}}$ mass estimated at the final step, the Pu_{KMP} mass can be calculated using Eq. (2). The systematic and random uncertainties of each term in Eq. (2) have been studied by KAERI [5]. A mere uncertainty propagation presented by KAERI would not be enough. A new scheme to evaluate the probability of Type-I error (α , false positive error) for the Pu MUF estimation, which is a performance metric for the safeguards assessment [16,17] has been developed by Woo et al. [18]. In the scheme proposed by Woo et al. [15], the α value was evaluated by the hypothesis testing method for representative used nuclear fuel assemblies assuming the Pu-to- ^{244}Cm ratio (see Eq. (2)) method recommended for Pu accounting. However, Woo et al.'s scheme of estimating Type-I error needs modification for analyzing Pu diversion scenarios in a pyroprocessing system. Modified Type-I error estimation as well as the associated sensitivities in Type-II error (β , false negative error) estimation for Pu material accounting using the Pu-to- ^{244}Cm ratio under hypothetical Pu diversion scenarios for a pyroprocessing facility are presented in this study.

2. Methodology

2.1. Loss of Pu ($LO\text{Pu}$)

A new random variable named Loss of Pu ($LO\text{Pu}$) is defined as the difference between the original Pu mass (Pu_O) with no Pu diversion and the Pu mass after diversion has occurred (Pu_L) as shown below,

$$LO\text{Pu} = \text{Pu}_O - \text{Pu}_L \quad (3)$$

Eq. (3) can be rewritten by substituting Eq. (2),

$$LO\text{Pu} = (\text{Ratio}_O \times {}^{244}\text{Cm}_O) - (\text{Ratio}_L \times {}^{244}\text{Cm}_L) \quad (4)$$

where Ratio_O and ${}^{244}\text{Cm}_O$ are the original Pu-to- ^{244}Cm ratio and ${}^{244}\text{Cm}$ mass, and Ratio_L and ${}^{244}\text{Cm}_L$ are the Pu-to- ^{244}Cm ratio and ${}^{244}\text{Cm}$ mass for the diversion scenario. The two random variables, Pu_O and Pu_L , are measured values using the Pu-to- ^{244}Cm ratio method. The Pu-to- ^{244}Cm ratio is a random variable consisting of a ratio of two normally distributed random variables, Pu and ^{244}Cm masses. The Pu-to- ^{244}Cm ratio of two independent Gaussian random variables does not follow a Gaussian distribution instead it follows the Cauchy distribution. However, the Geary-Hinkley transformation can be used to convert the ratio that follows a Cauchy distribution to a Gaussian distribution [19]. The approximate mean and variance of the transformed Gaussian distribution of the Ratio can be evaluated by solving the quadratic equation,

$$\begin{aligned} & \text{Ratio}^2 (\mu_{\text{Pu}}^2 - z^2 \sigma_{\text{Pu}}^2) - 2\text{Ratio}(\mu_{\text{Pu}}\mu_{244\text{Cm}}) + (\mu_{244\text{Cm}}^2 - z^2 \sigma_{244\text{Cm}}^2) \\ & = 0 \end{aligned} \quad (5)$$

where μ_{Pu} and $\mu_{244\text{Cm}}$ are the means of Pu and ^{244}Cm , σ_{Pu} and $\sigma_{244\text{Cm}}$ are the standard deviations of Pu and ^{244}Cm from their respective individual Gaussian distributions. The variable, z , is the critical value for the confidence interval. The z value is set as 1.96 for a 95% confidence interval in this study. The mean of random variable, Ratio , is the average of the two solutions of Eq. (5). The standard deviation of Ratio is the sum of two solutions of Eq. (5) divided by z . Then, the random variables, Pu_O and Pu_L can be represented by $\text{Pu}_O \sim N(\mu_{\text{Pu}_O}, \sigma_{\text{Pu}_O}^2)$ and $\text{Pu}_L \sim N(\mu_{\text{Pu}_L}, \sigma_{\text{Pu}_L}^2)$. The random variable, $LO\text{Pu}$, can also follow the normal distribution expressed as $LO\text{Pu} \sim N(\mu_{LO\text{Pu}}, \sigma_{LO\text{Pu}}^2)$. In this study, the uncertainties of measuring ^{244}Cm by using the coincidence neutron detection system denoted by ${}^{244}\text{Cm}_O$ and ${}^{244}\text{Cm}_L$ in Eq. (4) are ignored to focus on the effect of non-uniform Pu and ^{244}Cm spatial compositions in the uncertainty of evaluating the Pu-to- ^{244}Cm ratio on the NMA. The expected value and standard deviation for $LO\text{Pu}$ can be formulated as follows,

$$\mu_{LO\text{Pu}} = \mu_{\text{Ratio}_O} \times {}^{244}\text{Cm}_O - \mu_{\text{Ratio}_L} \times {}^{244}\text{Cm}_L \quad (6)$$

$$\sigma_{LO\text{Pu}} = \sqrt{(\sigma_{\text{Ratio}_O} \times {}^{244}\text{Cm}_O)^2 + (\sigma_{\text{Ratio}_L} \times {}^{244}\text{Cm}_L)^2} \quad (7)$$

In this study, a protracted diversion scenario is assumed, that is a few hundreds of grams of Pu is diverted per day for about a month (which is one MBP) to obtain one significant quantity (SQ) of 8 kg Pu. Details of the protracted diversion are shown in Table 1. After processing identical and independent multiple N used fuel assemblies, the expectation value (μ) and standard deviation (σ) of $LO\text{Pu}$ are evaluated by the properties of error propagation using

$$\begin{aligned} \mu_{LO\text{Pu}}^N &= \mu_{LO\text{Pu},1} + \mu_{LO\text{Pu},2} + \mu_{LO\text{Pu},3} + \cdots + \mu_{LO\text{Pu},N} = \mu_{LO\text{Pu}} \times N \\ & \quad (8) \end{aligned}$$

$$\begin{aligned} \sigma_{LO\text{Pu}}^N &= \sqrt{(\sigma_{LO\text{Pu},1})^2 + (\sigma_{LO\text{Pu},2})^2 + (\sigma_{LO\text{Pu},3})^2 + \cdots + (\sigma_{LO\text{Pu},N})^2} \\ &= \sigma_{LO\text{Pu}} \sqrt{N} \end{aligned} \quad (9)$$

Equations (7) and (8) are valid, if individual $\mu_{LO\text{Pu}}$ and $\sigma_{LO\text{Pu}}$ satisfy the following two conditions,

$$\begin{aligned} \mu_{LO\text{Pu},1} &= \mu_{LO\text{Pu},2} = \mu_{LO\text{Pu},3} = \cdots = \mu_{LO\text{Pu},N} = \mu_{LO\text{Pu}} \text{ and } \sigma_{LO\text{Pu},1} \\ &= \sigma_{LO\text{Pu},2} = \sigma_{LO\text{Pu},3} = \cdots = \sigma_{LO\text{Pu},N} = \sigma_{LO\text{Pu}} \end{aligned}$$

These two conditions are true in this study because every fuel assembly of same type is modeled identically. In a real scenario, these $\mu_{LO\text{Pu}}$ and $\sigma_{LO\text{Pu}}$ values for each assembly may not be exactly same.

2.2. Hypothesis testing

A null (H_0) and an alternative hypothesis (H_a) are defined where the random variable $LO\text{Pu}$ is respectively equal to zero and 8 kg. One SQ given by IAEA for Pu is 8 kg [1]. Since the $LO\text{Pu}$ cannot be 8 kg in one used fuel assembly the variable N in Eqs. (8) and (9) are calculated as the number of used fuel assemblies for which $LO\text{Pu}$ is equal to 8 kg. The probability of Type-II error (β) for $LO\text{Pu}$ can be then formulated as,

Table 1
Pu masses in used fuel assemblies considered for the diversion scenarios analysis.

Fuel assembly type	Pu mass for scenarios [kg]				
	Original Pu mass	Scenario 1		Scenario 2	
		5 pieces per rod	Pu mass ⁺ diverted per day	10 pieces per rod	Pu mass ⁺ diverted per day
Type-0	4.91	4.84 (122)*	0.30	4.78 (61)*	0.61
Type-1	5.30	5.23 (113)*	0.33	5.16 (56)*	0.66
Type-2	5.32	5.25 (112)*	0.33	5.17 (56)*	0.66

⁺Assuming 4.6 fuel assemblies on an average is chopped per day [22].

*The number in bracket indicates the number of fuel assemblies needed to obtain one SQ for the given diversion scenario.

$$\beta = \text{Prob} \left\{ \frac{LOPu - \mu_{LOPu}^N}{\sigma_{LOPu}^N} \leq \frac{S - \mu_{LOPu}^N}{\sigma_{LOPu}^N} | H_a \right\} = \Phi \left\{ \frac{S - \mu_{LOPu}^N}{\sigma_{LOPu}^N} \right\} \quad (10)$$

where S is the threshold value. Therefore, the probability of detection for $LOPu$ when there is only a single KMP after treating N used fuel assemblies is

$$1 - \beta = \Phi \left\{ \frac{\mu_{LOPu}^N - S}{\sigma_{LOPu}^N} \right\}. \quad (11)$$

2.3. Uncertainty for evaluating the Pu-to-²⁴⁴Cm ratio

In a previous study, the mean and standard deviation of the ratio, μ_{Ratio} and σ_{Ratio} , caused by the non-uniformity of nuclide composition in used fuel assemblies has been evaluated [18]. The tasks for evaluation in that study were as follows; 1) using fuel depletion simulations obtain the radial and axial non-uniformity Pu and ²⁴⁴Cm compositions in each fuel rod in three representative used fuel assemblies as shown in Figs. 1 and 2) modify these composition data from the simulations to depict the processing for materials in the chopping, voloxidation and granulation processes; 3) estimate the mean and standard deviation of Pu and ²⁴⁴Cm in the single granule as a function of powder and granule sizes; 4) substitute the mean and standard deviation values in the quadratic equation for the Geary-Hinkley transformation; and 5) assign the average of two solutions from the task-4 as the mean of the Pu-to-²⁴⁴Cm ratio and average of two solutions divided by the critical value as its standard deviation. The three nuclear fuel assemblies shown in Fig. 1 were selected because these three assembly types

were used in an equilibrium core of optimized power reactor-1000 [20,21], which is one of the representative reactor types in the Republic of Korea.

3. Evaluation for the probability of detection

3.1. Analysis of Pu material missing scenarios

As discussed in section 2.3, the fuel depletion simulation results using the SERPENT code are used to create the data (Table 2 in reference [18]) needed for the analysis of Pu material missing scenarios for a pyroprocessing facility. Among several nuclide composition data in that paper, the 3 cycles depletion results for Typ-0, Type-1 and Type-2 fuel assemblies are utilized in this study. The Pu masses in each chopped fuel rod piece, powder and granule can be obtained from this data. In order to develop Pu material diversion scenarios, it is assumed that material is diverted from chopped pieces of fuel rods (1 cm in length) right after the chopping process, which is the first process in the head-end process of pyroprocessing. The two different Pu material diversion scenarios analyzed are:

- Scenario 1: diversion of 5 pieces per rod,
- Scenario 2: diversion of 10 pieces per rod.

One fuel rod on an average is chopped into 381 pieces because the active fuel length is 381 cm. One fuel assembly consists of 236 fuel rods. Therefore, the total generated pieces by the chopping process for one fuel assembly is 89,916 pieces. Number of diverted pieces for two cases are 1180 (scenario 1), and 2360 (scenario 2) pieces per fuel assembly. The chopped pieces diverted are intentionally selected pieces originally located in the middle of fuel rods, because the Pu mass in the pieces from the middle part of the fuel

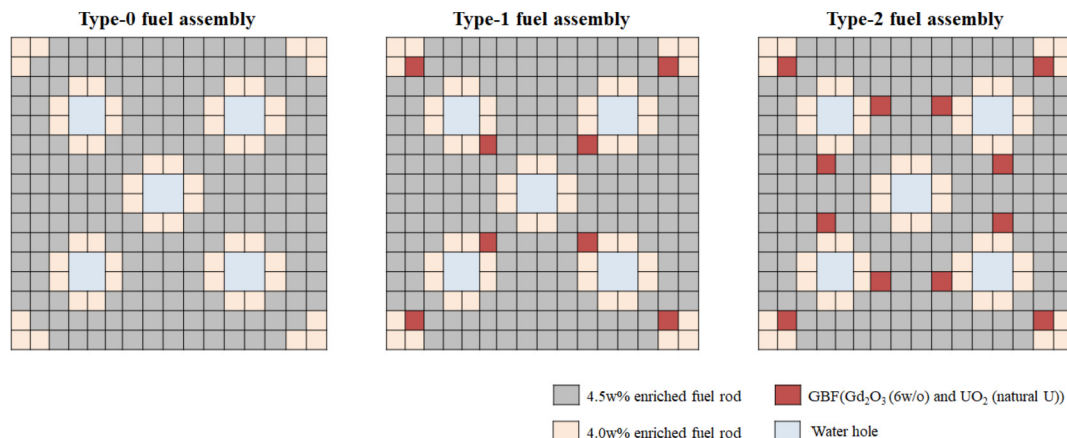


Fig. 1. Three representative fuel assembly types.

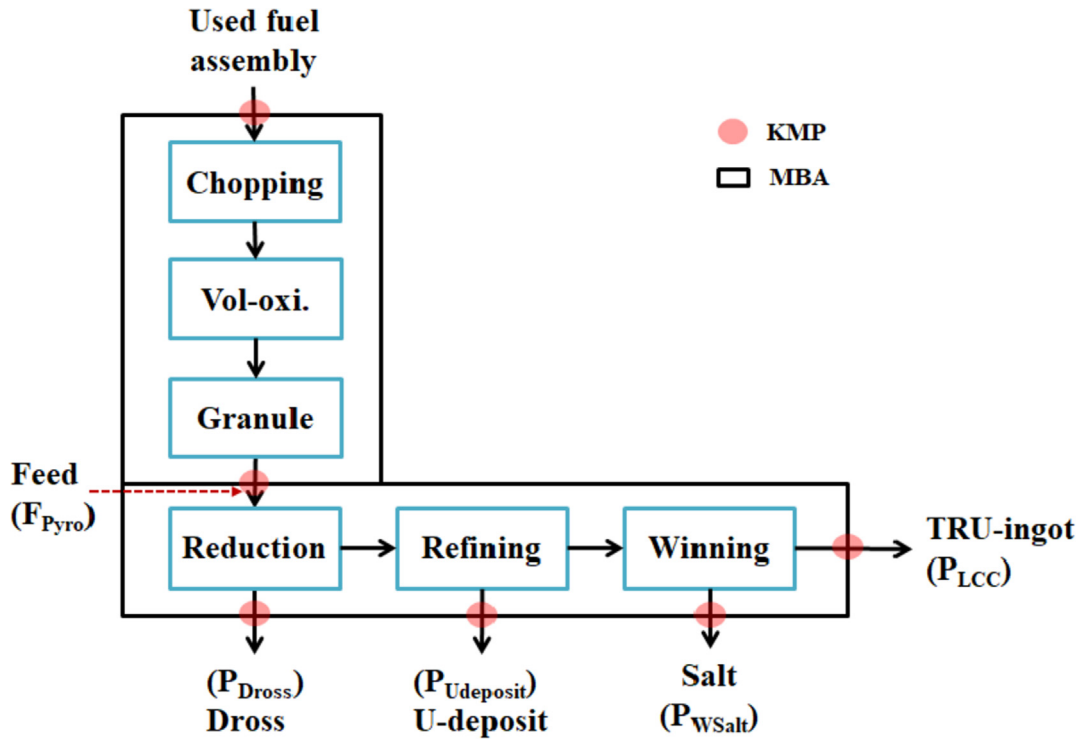


Fig. 2. The MBA-I for the key-pyroprocess.

rods is generally greater than those in the pieces from other locations. For example, in the case of scenario 1, the diverted pieces are from locations at the heights of 190 cm, 189 cm, 188 cm, 187 cm, and 186 cm from the bottom of the fuel rod. After the diversion has occurred, the remaining Pu masses that will be accounted for are summarized in Table 1. Table 1 also contains the data on original Pu masses if the diversion had not occurred. The column named 'Original Pu mass' in the table is the original Pu mass without involving any diversion. Then, the next two columns show the Pu mass for the diversion scenarios of 5 and 10 pieces per rod, respectively. The difference between the original Pu mass and the Pu mass after diverting from one single assembly is less than 1SQ of Pu. In this study, the objective is to consider the diversion of 8 kg of $LOPu$ in both scenarios. Hence, several assemblies are needed for each scenario (5 or 10 pieces per rod) for the $LOPu$ to be 8 kg. The number of assemblies should be integer; therefore, a ceiling function is applied to the count and number of assemblies, N is shown in the brackets in Table 1. The data of N is utilized in Eqs. (8)–(11).

There is no information available on the interval length of a MBP in a commercial pyroprocessing facility. However, literature from KAERI suggests that 4.6 fuel assemblies can be treated per day [22] which means the largest number of fuel assemblies from Table 1 (122) could be treated in one month which could become the MBP for the NMA. In addition to that, in a pyroprocessing facility, the product ingot (TRU-U ingot) falls under the IAEA category of "unirradiated direct use material" for which the suggested MBP is one month.

3.2. MBA for the key-pyroprocessing

The single MBA type (MBA-I) consisting of five KMPs including three main processes (the electrolytic-reduction, electro-refining, and electro-winning processes) as shown in Fig. 2 has been utilized in the previous study [18]. After completing the electrochemical process reaction, the dross floating above the molten salt solution

contains rare earth elements. This dross is mechanically separated and treated as waste [23]. U is recovered by the electro-refining process. After this process, the molten salt contains a small amount of U remainder and TRU elements. These elements are recovered together by a liquid cadmium cathode (LCC) of the electro-winning process. Then, products of the electro-winning process are manufactured to a TRU-U ingot, which is in the ratio 1:1. The key-pyroprocessing has several KMPs which are sequentially connected before and after each step. At each KMP there are two possible outcomes which are detection and non-detection of nuclear material diversion. If an event is detected at one KMP, it can be determined that this event is detected, even though the previous KMPs did not detect the event. Various possible outcomes of a given initiating event at each KMP can be expected and developed by the inductive logic method, the event tree method [24]. Based on the MBA shown in Fig. 2, the probability of detection (D) for the entire system can be computed by the event tree as shown in Fig. 3. Among 6 possible outcomes, the pathway for non-detecting an event is that the pathway consisting of non-detecting an event at all KMPs which is represented by 'ND' in the outcome column in Fig. 3. Except for this pathway, all other possible pathways can be judged to detect an event. One possible pathway consists of single or multiple components of detection or non-detection. These multiple components are connected in series, therefore, the probability of detection for one possible pathway is the multiplication of detection or non-detection probabilities at each KMP. Each possible pathway for detecting an event is independent. Therefore, the total probability of detection (TDP) for the $LOPu$ event in the system is the summation of probabilities of detection for each possible pathway in the MBA-I as formulated,

$$TDP = \sum_{i=1}^5 \text{Prob}\{D_i\} \quad (12)$$

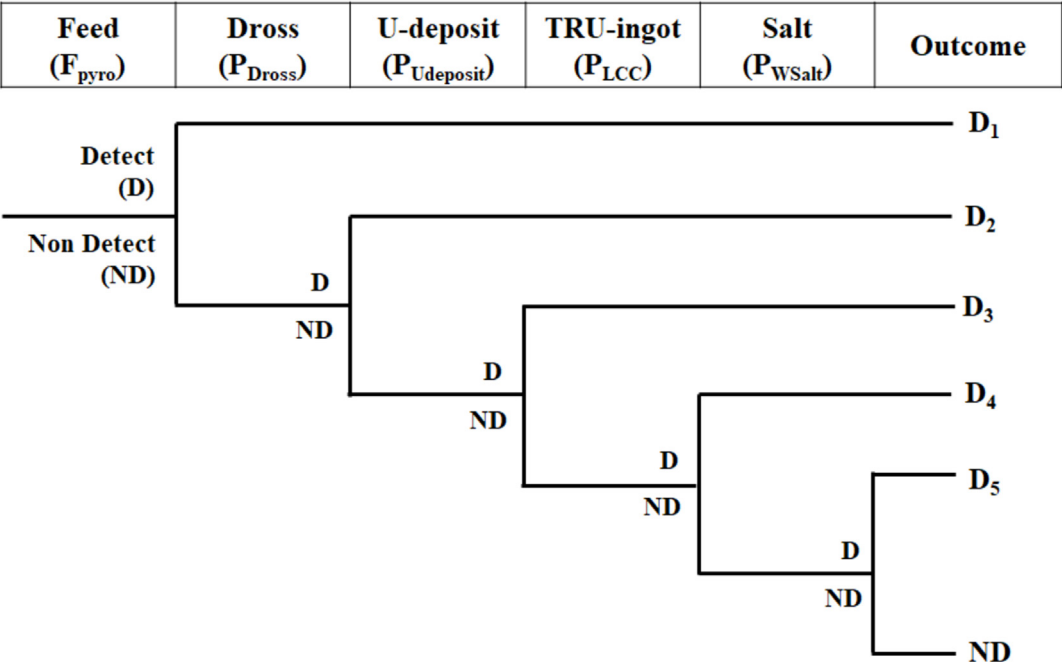


Fig. 3. Event tree for the MBA-I.

where $\text{Prob}\{D_i\}$ is the probability of detection in the i^{th} -pathway.

In a previous study, depending on the size of powder and granule sampled, the uncertainty for evaluating the Pu-to- ^{244}Cm ratio associated with the non-uniformity of nuclide composition in representative used nuclear fuel assemblies was evaluated [18]. The uncertainty of this ratio, which is mathematically a result from Eq. (5), as a function of power and granule size is substituted into Eq. (11) with the N values shown in the brackets of Table 1. Then, the probability of detection is evaluated using Eq. (12). Finally, Fig. 4

shows the probability of detection for the LOPu as a function of the radii of the granule and the powder particle sizes for both scenarios of Pu diversion when i) the used fuel assembly is of Type-0, Type-1 and Type-2 after 3 cycles of fuel depletion and ii) a single granule is randomly selected as a sample. The current suggestion for material accountancy is the probability of detection should be greater than or equal to 95%, which is corresponding to the dark orange color area in Fig. 4. From Fig. 4, it is seen that there is a high possibility of probability of detection for the LOPu to be less than

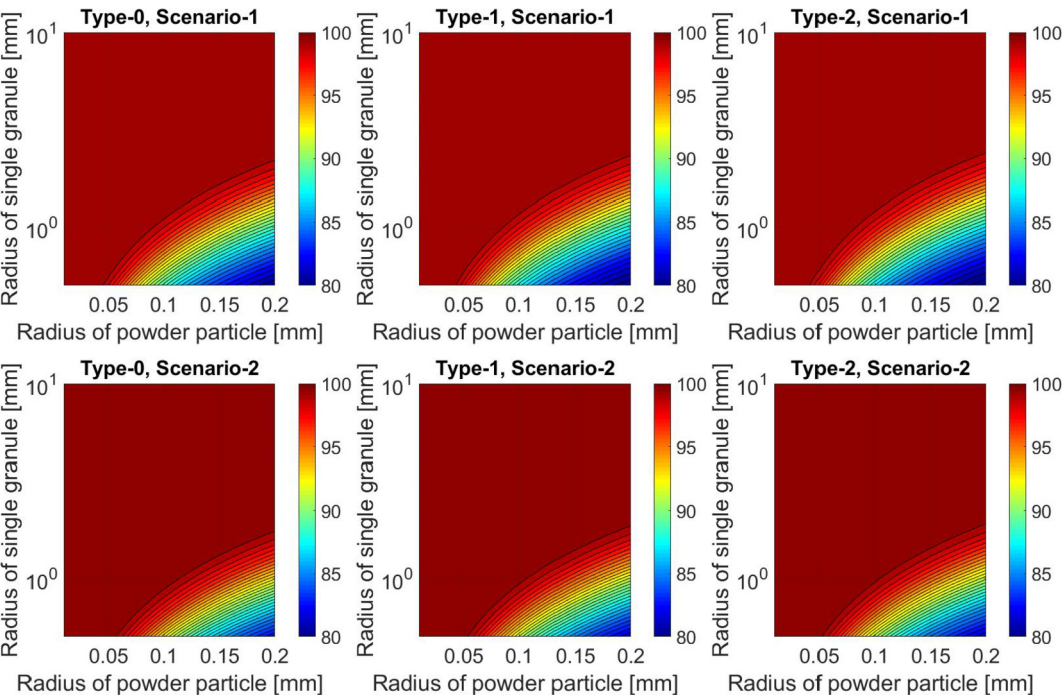


Fig. 4. Total probability of detection for the LOPu in the key-pyroprocess for the Type-0, Type-1, and Type-2 assembly after 3 cycles depletion under scenarios 1 and 2.

95% when a single granule is sampled to evaluate to the ratio of Pu-to- ^{244}Cm . The area for probability of detection greater than 95% is getting greater as the number of missing pieces increase (scenario 1 to 2). This is because it is easier to detect larger amounts of diverted material. These results show how the non-uniformity of nuclide composition in used nuclear fuel assemblies affect the detection of loss of Pu scenario. As shown in the previous study, increasing the samples size (by increasing the number of granules in the sample) significantly enhanced the material accountability limited by the non-uniformity of nuclide composition in used nuclear fuel assemblies [18]. In the next section, change of *TDP* for the Pu diversion scenario are analyzed with respect to the reconfiguration of the MBA and due to the increase in sample size.

3.3. New MBA model for the key-pyroprocessing

The new MBA type (MBA-II) for the key-pyroprocessing is modeled in this section. The main concept is that each batch process in the key-pyroprocessing is set as an independent MBA. Therefore, the key-pyroprocessing consists of three individual MBAs as shown in Fig. 5. Then, the event tree is built to evaluate the probability of detection for the *LOPu* condition in the system as shown in Fig. 6. The order of KMPs in the event tree is based on the order of mass flow stream from the electrolytic-reduction to the electro-winning process shown in Fig. 5. Since more number of KMPs are employed in the MBA-II model, more outcomes are possible compared to that of the MBA-I model shown in Fig. 3. The probability of detection for each pathway is evaluated by taking the product of probability of detection or non-detection in that pathway. Finally, the *TDP* in the MBA-II system can be evaluated as the summation of all detecting scenario pathways as,

$$TDP = \sum_{i=1}^7 \text{Prob}\{D_i\} \quad (13)$$

In order to compare the probability of detection for the *LOPu* event between the two MBA types, the Type-2 assembly after the 3 cycles of depletion is selected for scenario 1 only. The *TDP* for the MBA-I (left) and MBA-II (right) are plotted as a function of powder and granule size when a single granule is randomly selected as a sample in Fig. 7. The two contour plots in Fig. 7 show the effect on *TDP* due to the increasing in the number of MBAs or KMPs in the system. The color-map range for the MBA-I model is widely spread out from 80% to 100%, in contrast, the *TDP*s of the 8 kg *LOPu* event for the MBA-II model are mostly greater than 96%, even though only single granule is taken as a sample to evaluate the ratio in the head-end process. The dramatic increase in the probability of detection is attributed to the large number of KMPs because it increases the chances for detecting unanticipated or undesirable events. The increasing the number of KMPs can enhance the NMA, on the other hand, it could also increase the time for safeguards, processing, and operating the system. These concepts and approaches could be utilized as a methodology to design the MBA for the nuclear facilities.

All previous results are based on the sample size of granule being one. However, in practice the sample size can vary. In order to investigate the effect of varying sample size, the probability of detection for the 8 kg *LOPu* using the Type-0 used fuel assembly after the 3 cycles depletion when one granule is taken as a sample is compared to the results when the number of granule sample is ten and results are shown in Fig. 8. The result for the ten granules case shows significant increase in the area for probability of detection greater than 95%, although the enhancement of detection probabilities is not as much for the case where MBA model is changed from MBA-I to MBA-II.

Hence, it can be concluded that increasing the number of MBAs along with KMPs and the sample sizes can avoid the protracted Pu diversion events. This is due to the possibility of including in the calculations the uncertainty associated with the non-uniformity of Pu and ^{244}Cm nuclide composition. Moreover, the number of

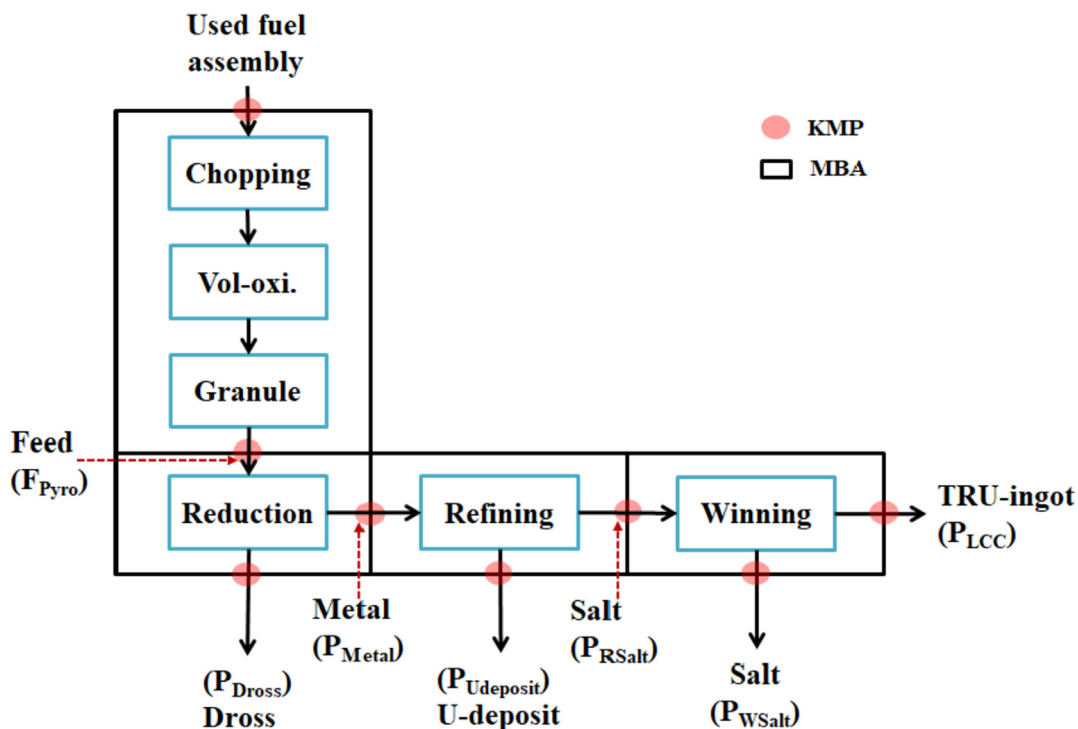


Fig. 5. The new MBA type (MBA-II) for the key-pyroprocess.

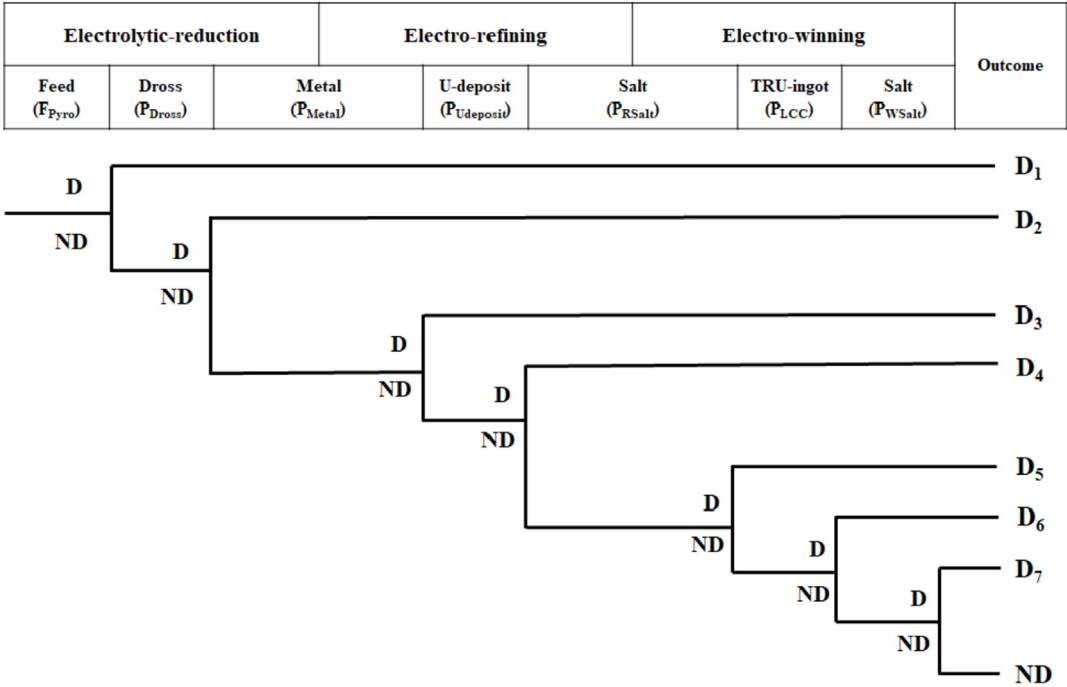


Fig. 6. Event tree for evaluating the total probability of detection for the *LOPu* in the new MBA type.

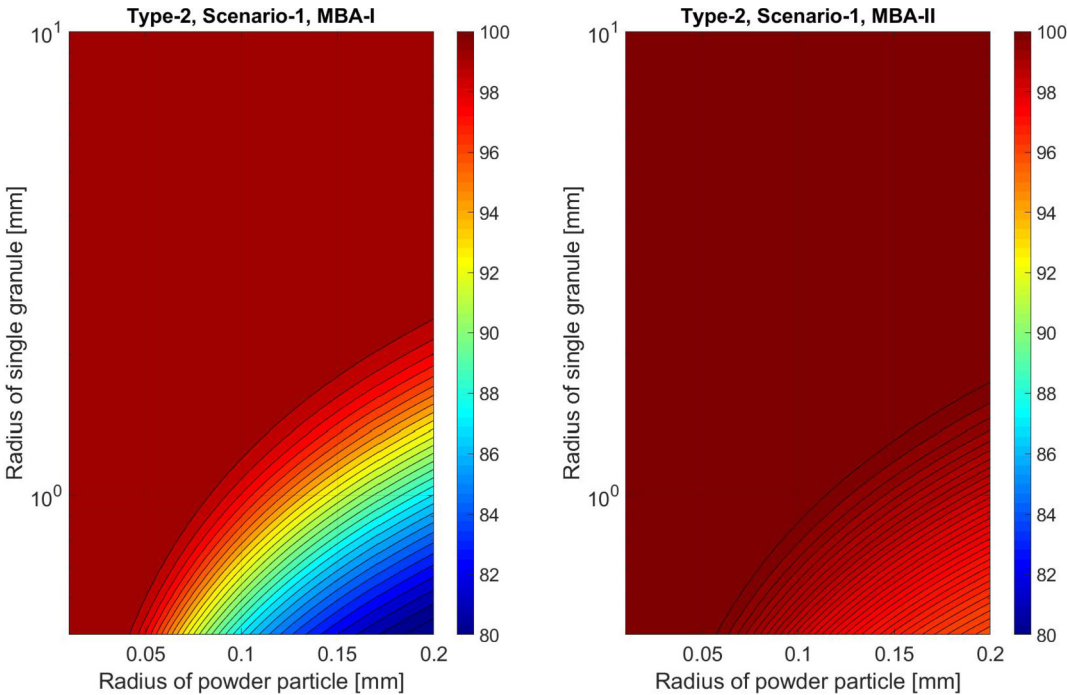


Fig. 7. Probability of detection for the *LOPu* in the key-pyroprocess for Type-1 fuel assembly based on two MBA models (left: the MBA-I model, and right: the MBA-II model) when the missing 5 pieces per rod case is applied.

assemblies necessary for the *LOPu* to be 8 kg for the Pu diversion scenario 1 is approximately 110~150 as shown in Table 1. According to the conceptual design for Korea Advanced pyroprocessing Facility Plus, around 4.6 spent fuel assemblies are planned to be treated per day. At that rate, it would take about 20~30 days for processing of 110~150 assemblies. After every campaign, inspection should be conducted by both operators and inspectors. Those inspection and verification works avoid the *LOPu* getting near 8 kg.

This approach would be supportable to decide the MBP of pyroprocessing.

4. Summary and conclusion

A new methodology to evaluate the Pu mass accountability in pyroprocessing is developed. Among several uncertainty sources of material accounting in used fuel pyroprocessing, this study focused

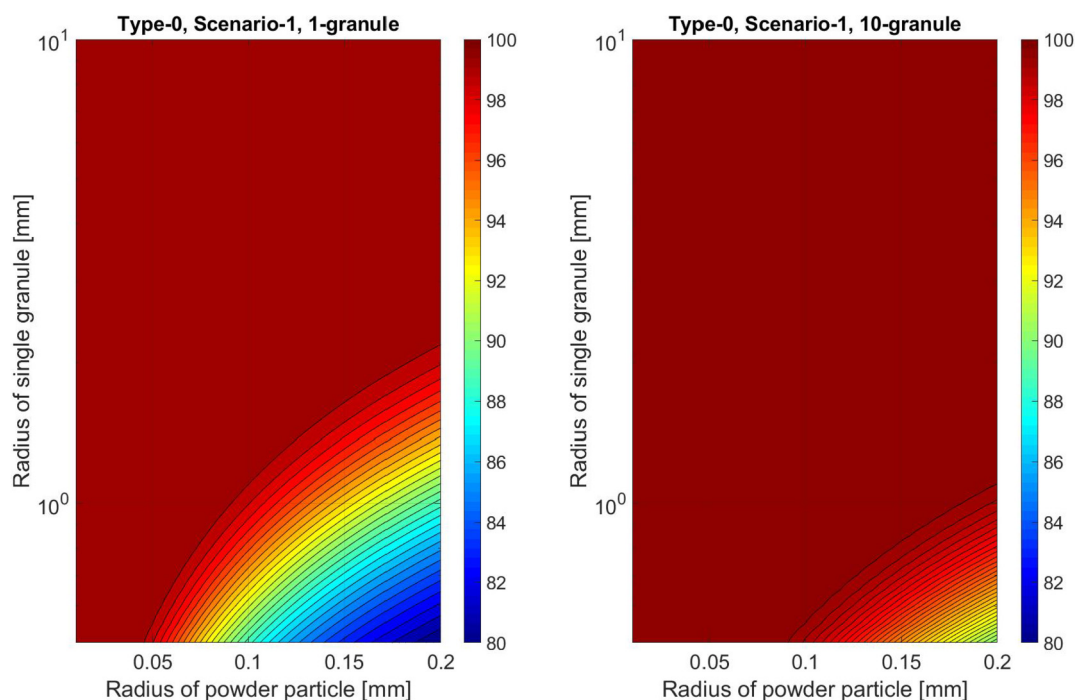


Fig. 8. Probability of detection for the $LOPu$ in the key-pyroprocess for Type-0 fuel assembly in the original MBA model (Fig. 2) under the missing 5 pieces per rod case when the sample size of granule for evaluating the ratio is one (left) and ten (right).

on how the non-uniform nuclide composition of Pu and ^{244}Cm in used fuel assemblies could affect the detection of Pu material loss if a planned Pu-to- ^{244}Cm ratio is used for this purpose. To test the feasibility of the new method proposed, results of a previous study consisting of high fidelity reactor core physics and fuel depletion simulations are utilized. These simulation results provided the radial and axial spatial distributions of Pu and ^{244}Cm for each used fuel rod of representative fuel assemblies. Moreover, available from that study were the uncertainties in evaluating the Pu-to- ^{244}Cm ratio for Type-0, -1, and -2 representative used fuel assemblies. Based on the previous study aforementioned a random variable, $LOPu$, is newly defined as the difference between the original Pu mass and the Pu mass after the protracted diversion of 1SQ Pu material. The probability of detection for the 8 kg $LOPu$ in the pyroprocessing system is evaluated with respect to the size of granule and powder using the hypothesis testing method and the event tree analysis. It is observed that for some of the hypothetical Pu material diversion scenarios the probability of detection for the 8 kg $LOPu$ is less than 95%. In order to enhance the probability detection, a new MBA scheme with more number of KMPs for the key-pyroprocess is proposed and analyzed. The probabilities of detection for used fuel assemblies based on this new scheme is found to be greater than 96%. It is also observed that when the granule sample size increases from one to ten, the probability of detection for the $LOPu$ significantly increased. Based on these observations, even though the material accountability could be affected by the non-uniformity of nuclide composition, it can be concluded that this weakness could be surmounted by decreasing the uncertainty of the ratio by increasing the sample size and by modestly increasing the number of MBAs and number of KMPs in the pyroprocessing system.

The robust NMA for pyroprocessing that utilizes the MBA-II configuration presented in this study and an increased sample size (i.e. number of granules per sample) can alleviate the proliferation risk. The provision to use the uncertainty in the Pu-to- ^{244}Cm due to the non-uniformity of Pu and ^{244}Cm nuclide

composition presented in this study is also helpful for making the NMA robust. In the future, additional sources of uncertainty, such as instrument uncertainties, will be considered along with the Pu and ^{244}Cm nuclide composition uncertainties considered in this study.

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References

- [1] International Atomic Energy Agency (IAEA), IAEA Safeguards Glossary 2001 Edition, 2001.
- [2] Nuclear Material Accounting Handbook, IAEA Services Series No. 15, 2008.
- [3] P.C. Durst, R. Wallace, I. Therios, M.H. Ehinger, R. Bean, D.N. Kovacic, A. Dougan, K. Tolks, B. Boyer, Advanced Safeguards Approaches for New Reprocessing Facilities, 2007.
- [4] R. Avenhaus, Material Accountability: Theory, Verification, and Applications, John Wiley & Sons, Chichester, UK, 1977.
- [5] B. Han, H. Shin, H. Kim, Analysis of measurement uncertainty of material unaccounted for in the reference pyroprocessing facility, Nucl. Technol. 182 (2013).
- [6] H.L. Chang, F.X. Gao, W.I. Ko, H.D. Kim, S.Y. Lee, Evaluation of sigma-MUF (Material Unaccounted For) for the conceptually designed Korea advanced pyroprocess facility, J. Kor. Phys. Soc. 59 (2011) 1418, <https://doi.org/10.3938/jkps.59.1418>.
- [7] N. Miura, H.O. Menlove, The Use of Curium Neutrons to Verify Plutonium in Spent Fuel and Reprocessing Wastes, 1994.
- [8] T.-H. Lee, Y.-S. Kim, T.-J. Kwon, H.-S. Shin, H.-D. Kim, Determination of the plutonium mass and curium ratio of spent fuel assemblies for input nuclear material accountability of pyroprocessing, and analysis of their errors, Nucl. Technol. 179 (2012) 196–204.
- [9] M. Gonzalez, L. Hansen, D. Rappleye, R. Cumberland, M.F. Simpson, Application of a one-dimensional transient electrorefiner model to predict partitioning of plutonium from curium in a pyrochemical spent fuel treatment process, Nucl. Technol. 192 (2015) 165–171, <https://doi.org/10.13182/NT15-28>.
- [10] H. Lee, G. Il Park, J.W. Lee, K.H. Kang, J.M. Hur, J.G. Kim, S. Paek, I.T. Kim, I.J. Cho, Current status of pyroprocessing development at KAERI, Sci. Technol. Nucl. Install 2013 (2013) 1–11.
- [11] T.H. Lee, H.O. Menlove, S.Y. Lee, H.D. Kim, Development of the ACP safeguards

- neutron counter for PWR spent fuel rods, Nucl. Instrum. Methods Phys. Res., Sect. A 589 (2008) 57–65, <https://doi.org/10.1016/j.nima.2008.02.054>.
- [12] T.H. Lee, H.D. Kim, J.S. Yoon, S.Y. Lee, M. Swinhoe, H.O. Menlove, Preliminary calibration of the ACP safeguards neutron counter, Nucl. Instruments methods Phys. Res. Sect. A Accel. Spectrometers, Detect. Assoc. Equip 580 (2007) 1423–1427, <https://doi.org/10.1016/j.nima.2007.07.142>.
- [13] T.H. Lee, Y.S. Kim, H.-S. Shin, H.-D. Kim, Hot-test results of the advanced spent fuel conditioning process safeguards neutron counter for PWR spent fuel rods, Nucl. Technol. 176 (2011) 147–154, <https://doi.org/10.13182/NT11-A12549>.
- [14] T.H. Lee, H.D. Kim, Application of a self-multiplication correction method to a neutron coincidence counter and its calibration for spent fuel, IEEE Trans. Nucl. Sci. 56 (2009) 2791–2795, <https://doi.org/10.1109/TNS.2009.2021427>.
- [15] H. Seo, B.H. Won, S.K. Ahn, S.K. Lee, S.H. Park, G. Il Park, S.H. Menlove, Optimization of hybrid-type instrumentation for Pu accountancy of U/TRU ingot in pyroprocessing, Appl. Radiat. Isot. 108 (2016) 16–23, <https://doi.org/10.1016/j.apradiso.2015.11.109>.
- [16] R.A. Borrelli, Use of curium spontaneous fission neutrons for safeguardability of remotely-handled nuclear facilities: fuel fabrication in pyroprocessing, Nucl. Eng. Des. 260 (2013) 64–77, <https://doi.org/10.1016/j.nucengdes.2013.03.025>.
- [17] R.A. Borrelli, Functional components for a design strategy: hot cell shielding in the high reliability safeguards methodology, Nucl. Eng. Des. 305 (2016) 18–27, <https://doi.org/10.1016/j.nucengdes.2016.05.010>.
- [18] S.M. Woo, S.S. Chirayath, M. Fratoni, Nuclide Composition Non-uniformity in Used Nuclear Fuel for Considerations in Pyroprocessing Safeguards, 2018. Nucl. Eng. Technol. (n.d.), <https://doi.org/10.1016/j.net.2018.05.011>.
- [19] J. Hayya, D. Armstrong, N. Gressis, A note on the ratio of two normally distributed variables, Manag. Sci. 21 (1975) 1338–1341, <https://doi.org/10.1287/mnsc.21.11.1338>.
- [20] H. Yu, Low-Boron OPR1000 Core Based on the BigT Burnable Absorber, KAIST, 2014.
- [21] H. Yu, M.S. Yahya, Y. Kim, A reduced-boron OPR1000 core based on the BigT burnable absorber, Nucl. Eng. Technol. 48 (2016) 318–329, <https://doi.org/10.1016/j.net.2015.12.010>.
- [22] S.K. Kim, W.I. Ko, S.R. Youn, R. Gao, Cost analysis of a commercial pyroprocess facility on the basis of a conceptual design in Korea, Ann. Nucl. Energy 80 (2015) 28–39.
- [23] J.-H. Yoo, C.-S. Seo, E.-H. Kim, H.-S. Lee, A conceptual study of pyroprocessing for recovering actinides from spent oxide fuels, Nucl. Eng. Technol. 40 (2008) 581–592, <https://doi.org/10.5516/NET.2008.40.7.581>.
- [24] N.J. McCormick, Reliability and Risk Analysis: Methods and Nuclear Power Applications, Academic Press, New York, 1981.