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## ARTICLE

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### Safeguards approach and design of transuranium fuel cycle with accelerator-driven system based on material attractiveness

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The material attractiveness of plutonium (Pu) was evaluated for nuclear non-proliferation by assuming the diversion of items from the facilities in the transuranium fuel cycle with an accelerator-drive system (ADS cycle). The evaluation results were compared with the material attractiveness of mixed oxide (MOX) fuel assemblies for conventional boiling water reactors (BWRs). All items in the ADS cycle, regardless of whether they were fresh or spent fuel, were found to have the same attractiveness as Pu in the BWR-MOX spent fuel assembly. Additionally, to design and evaluate accounting systems for safeguards, the assumed uncertainty ( $\sigma_{\text{MUF}}$ ) values of measurements by operators and verification by inspectors for the Pu mass flow in the ADS cycle were derived with reference to the accuracy targets for Pu measurement technology and the attractiveness. Based on the discussion using the estimated  $\sigma_{\text{MUF}}$  and the target values defined by the significant quantity, we concluded that the appropriate inspection frequencies for the operator and inspector were almost 2 weeks and almost 5 days, respectively.

**Keywords:** material attractiveness; ADS; Pu; safeguards;  $\sigma_{\text{MUF}}$

#### 1. Introduction

Nuclear power generates electricity without emitting carbon dioxide, and is regarded as an important baseload electricity source. However, nuclear power plants still have the problem of disposing of high-level radioactive waste (HLW) in spent fuel. Research and development of the transuranium (TRU) fuel cycle with accelerator-drive systems (ADSs) transmuting minor actinides (MAs) separated from commercial cycles has been continuously conducted by the Japan Atomic Energy Agency (JAEA) to reduce the HLW contained in the spent fuel discharged from nuclear power plants [1-5]. ADSs are designed to operate on fuels in which MAs account for approximately 50–70 wt% of the actinide (An) composition to achieve efficient MA transmutation. The JAEA proposed a nitride with high stability for flexible An compositions as the fuel material and a pyrochemical process using liquid cadmium (Cd) as a reprocessing agent for the ADS spent fuel. The pyroprocessing products are generally less pure than those obtained by reprocessing using multistep solvent extraction. The mass balance evaluation result of the TRU fuel cycle with ADS (ADS cycle) was reveals that rare-earth elements (REs) are impurities accounting for 3.3 % of the fresh fuel produced by pyroprocessing [6].

Furthermore, the inspection goal of the safeguards

(SGs) required for the ADS cycle must be examined because the chemical form and composition of ADS fuels are different from those of current commercial fuels. However, the inspection goal for the SGs required for each facility in the ADS cycle have not been discussed, despite it being a unique nuclear system. Material unaccounted for (MUF) and its measurement relative uncertainty ( $\sigma_{\text{MUF}}$ ) evaluations are very important in reporting measurement results for accounting systems, which are the basis of SGs, to inspectors such as governments and the International Atomic Energy Agency (IAEA). MUF refers to the quantity that cannot explain the nonzero balance in the material balance area (MBA), which, in principle, should be zero. The diversion of MUF for purposes other than declarations is a great concern for inspectors. This concern can be addressed by  $\sigma_{\text{MUF}}$ , which is calculated as the uncertainty of the overall mass balance within each MBA, by accumulating the individual measurement uncertainties for the accounting system. The validity of the MUF is evaluated by comparison with this  $\sigma_{\text{MUF}}$ . If the measurement accuracy is low, the  $\sigma_{\text{MUF}}$  will be large. This result would be unacceptable to inspectors, as it would increase the MUF, which would be considered reasonable. In the ADS cycle as well, the  $\sigma_{\text{MUF}}$  target value must be considered in designing an SGs system that is acceptable to the inspectors.

This study focuses on a method for evaluating material attractiveness (herein referred to as “attractiveness”),

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which indicates the relative usability in theft for the purpose of manufacturing a nuclear explosive device (NED), as an index to examine the inspection goal required for the next-generation nuclear fuel cycle [7]. This method has a track record in being evaluated in nuclear fuel cycles including fast reactors and high-temperature gas-cooled reactors [7,8]. However, the evaluation has only been performed for the ADS facility within the ADS cycle [9]. The inspection goal required for the ADS cycle site can be discussed by evaluating the attractiveness of the other facilities in the ADS cycle.

In this study, attractiveness was evaluated considering the fundamental fuel property as an important factor for deciding the inspection goal required for the facility, assuming the diversion of fuel items from the reprocessing and fabrication complex (RFC) in the ADS cycle. Additionally, to design and evaluate accounting systems for SGs, the  $\sigma_{\text{MUF}}$  of measurements by operators and verification by inspectors for the plutonium (Pu) mass flow in the ADS cycle were derived with reference to the accuracy targets for Pu measurement technology and the attractiveness evaluation results. Finally, an acceptable inspection frequency was discussed by comparing the estimated  $\sigma_{\text{MUF}}$  values with their target values, based on the significant quantity (SQ) defined by the IAEA.

## 2. Attractiveness evaluation

### 2.1. Methodology and calculation conditions

We conducted the material attractiveness evaluation for nuclear non-proliferation for the inherent features of the ADS fuel in the RFC using the method developed by Aoki et al. [8], which was extended based on that for nuclear security developed by Bathke et al. [7]. Here, we assumed that the adversary would eventually manufacture an NED made of pure Pu (Pu-NED). In addition, we evaluated the attractiveness of mixed oxide (MOX) fuel assemblies stored in a commercial boiling water reactor (BWR) facility for the Pu-NED manufacturing case for comparison with that of the materials in the RFC in the ADS cycle. In this study, to evaluate the material attractiveness in terms of non-proliferation, the adversary

was assumed to be a state actor manufacturing Pu-NED from the diverted materials. For material attractiveness assessment for state actors, a proliferating state intending to divert the safeguarded nuclear material into Pu-NED was assumed to have the following characteristics: (1) having advanced technology, well-developed industries, and abundant capital; (2) having no natural U resources; (3) signatory the Non-Proliferation Treaty, comprehensive SGs agreement, and additional SGs agreements; (4) requiring 50% reliability for the NED; and (5) requiring the production of one NED [10,11]. In the evaluation for state actors, the physical properties of important nuclear materials were evaluated in each phase, i.e., processing and utilization, as shown in **Figure 1**. The conversion times defined by the IAEA [12] were used as indicators for evaluation in the processing phase. The indicators of the utilization phase were the bare critical mass (BCM) and heat content per BCM (HC). The BCM was calculated using the Monte Carlo calculation code MCNP-6.1.1 [13] together with the nuclear data library JENDL-4.0 [14]. The MCNP calculations used 1,050 neutron generations, with 500,000 histories per generation. The first 50 generations were excluded from the statistics for each case, yielding 500,000,000 active histories in each calculation. The standard deviation with respect to the multiplication factor is 0.03%. The HCs were obtained from calculations using the isotope generation and depletion calculation code ORIGEN2.2 [15] with the set of cross-section library ORLIBJ40 [16]. **Table 1** presents a categorization metric for the material attractiveness evaluation for state actors [7,8]. Finally, the lowest attractiveness level among the evaluation results was considered as the overall attractiveness of the theft target item.

### 2.2. Evaluation targets

As shown in **Figure 2**, the electrorefining step is carried out using a LiCl-KCl eutectic melt at 773 K to recover Ans separated from the fission products (FPs) in the pyroprocessing for the spent nitride fuel, which is based on the latest electrorefining design for the reprocessing of spent metal fuel [17]. A basket filled with spent nitride fuel

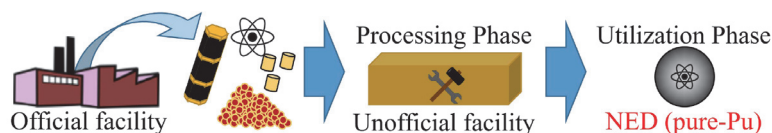


Figure 1. Processing and utilization phases.

Table 1. Categorization metric for material attractiveness evaluation for state actors [7,8].

| Attractiveness level | Processing phase                    | Utilization phase       |                            |
|----------------------|-------------------------------------|-------------------------|----------------------------|
|                      | Conversion time                     | BCM (kg) <sup>(a)</sup> | HC <sup>(a)</sup> (kW/BCM) |
| High (H)             | 1 week (Non-irradiated metal)       | <80                     | <0.45                      |
| Medium (M)           | 1–3 weeks (Non-irradiated compound) | 80–800                  | 0.45–4.5                   |
| Low (L)              | 1–3 months (Irradiated material)    | 800–4000                | >4.5                       |
| Very Low (VL)        | 3–12 months (Low enriched U)        | >4000                   |                            |

<sup>(a)</sup> BCM and HC are indices calculated under the assumption of the  $\alpha$  phase.

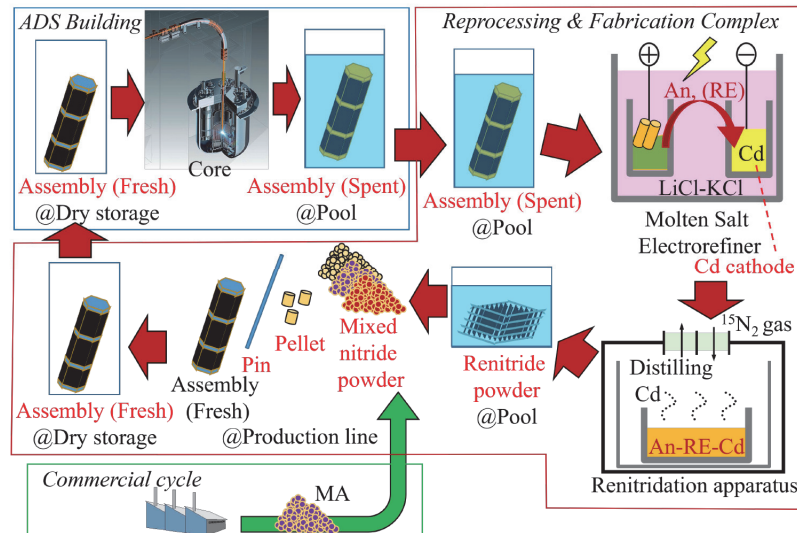


Figure 2. Item flow in the ADS cycle.

Table 2. Pu isotopic fraction [9].

| Nuclide | Mass fraction (wt.%)                               |  |  |  |
|---------|--|--|--|--|
|         | Fresh fuel of 1 <sup>st</sup> cycle <sup>(a)</sup> | Spent fuel of 1 <sup>st</sup> cycle <sup>(b)</sup> | Fresh fuel of 5 <sup>th</sup> cycle <sup>(a)</sup> | Spent fuel of 5 <sup>th</sup> cycle <sup>(b)</sup> |
| Pu-238  | 2.4  | 26.1   | 46.7   | 49.4   |
| Pu-239  | 54.7   | 35.7   | 17.3   | 15.0   |
| Pu-240  | 25.0   | 24.2   | 23.9   | 23.5   |
| Pu-241  | 10.9   | 5.8  | 2.5  | 2.3  |
| Pu-242  | 7.0  | 8.2  | 9.6  | 9.8  |

<sup>(a)</sup> For mixed nitride powder, pellet, pin, and assembly (fresh),<sup>(b)</sup> For assembly (spent), renitride powder, and Cd cathode.

serves as the anode. Liquid Cd is used as cathode to recover Ans. Ans recovered to the liquid Cd cathode by electrorefining are converted to nitride by heating in a  $^{15}\text{N}_2$  gas stream [18,19]. Ans renitride powder produced by the electrorefining is mixed with MA nitrides fabricated from partitioned MA generated in a commercial fuel cycle (mixed nitride powder) [20]. The mixed nitride powder is used as ADS fuel for the next burn-up cycle after being fabricated into pellets, pins, and assemblies.

In this study, the evaluation target items were Cd cathodes, including Ans, renitride powder, mixed nitride powder, pellet, pin, and assembly. The Pu compositions of the ADS burnup calculation results [9] were used to evaluate the material attractiveness of Pu. The isotopic ratios of  $^{238}\text{Pu}$  and  $^{239}\text{Pu}$  reached equilibrium after the 5<sup>th</sup> cycle, with no significant fluctuation. Thus, the Pu isotopic fractions of the 1<sup>st</sup> cycle of the launching stage and the 5<sup>th</sup> cycle in which the composition was in the equilibrium stage were the attractiveness evaluation targets of Pu-NED (Table 2). In addition, the MA and RE were always included in the fuel component during the ADS cycle. Thus, the removal of MA and RE is also a point of discussion in the processing phase.

### 2.3. Results

The evaluation results for material attractiveness for

nuclear non-proliferation are presented in Table 3. The evaluation results of the ADS fuel assemblies (fresh and spent) and BWR-MOX fuel assemblies (fresh and spent) used for comparison have been quoted from a previous study [9].

First, we discussed the evaluation results for the mixed nitride powder, pellet, pin, and assembly (fresh). The separation of nitrogen from nitride fuel was as complex as that of oxygen from MOX, despite the extraction of pure Pu from materials containing REs and MAs requiring more complex processing steps and longer processing times. Therefore, the conversion times of the mixed nitride powder, pellet, pin, and assembly (fresh) of the 1<sup>st</sup> and 5<sup>th</sup> cycles were categorized as attractiveness level L, which was the same as that of the BWR spent fuel. The BCMS of the mixed nitride powder, pellet, pin, and assembly (fresh) of the 1<sup>st</sup> and 5<sup>th</sup> cycles were categorized into the same level as those of the BWR fresh and spent fuels, regardless the small difference in the exact BCM value owing to  $^{238}\text{Pu}$ . The HCs of the mixed nitride powder, pellet, pin, and assembly (fresh) of the 1<sup>st</sup> cycle were categorized into attractiveness level H, which was the same level as that of the BWR fresh and spent fuels. However, the HC of the 5<sup>th</sup> cycle, was categorized as attractiveness level M, which was lower than that of the BWR fresh and spent fuels owing to the accumulation of  $^{238}\text{Pu}$  with large decay heat.

Second, we discussed the evaluation results of the assembly (spent), renitride powder, and Cd cathode. The assembly (spent) was categorized into attractiveness level L, which was the same level as the BWR spent fuels because it contained the FPs. The conversion times of the renitride powder including components obtained by the ADS spent fuel of the 1<sup>st</sup> and 5<sup>th</sup> cycles were categorized as attractiveness level L, which was the same level as the BWR spent fuel. Renitride powder is a product obtained after the extraction of FPs, but it still contains REs and MAs. The extraction of pure Pu from renitride powder containing REs and MAs requires more complex processing steps and longer processing times, similar to the spent fuel assemblies of ADS and BWR-MOX. The conversion times of the Cd cathode, including the components obtained by the ADS spent fuel of the 1<sup>st</sup> and 5<sup>th</sup> cycles were also categorized into the same attractiveness level as those of the BWR spent fuel. REs and MAs were recovered in the Cd cathode, while alkali metals (ALs) and alkaline earth metals (ALEs) in the FPs remained in the molten salt. Extracting pure Pu from a mixture of REs and Ans required a long processing time and complex processing steps, whereas the separation of Cd was relatively easy because of the difference in boiling points between Cd and the other elements. The BCMs of the assembly (spent), renitride powder, and Cd cathode of the 1<sup>st</sup> and 5<sup>th</sup> cycles were categorized into the same attractiveness level as those of the BWR fresh and spent fuels, despite the small difference in the exact BCM value owing to <sup>238</sup>Pu. However, the HCs of the assembly (spent), renitride powder, and Cd cathode of the 1<sup>st</sup> and 5<sup>th</sup> cycles were attractiveness level M, which was lower than the attractiveness levels of the BWR fresh and spent fuels owing to the accumulation of <sup>238</sup>Pu with a large decay heat.

Finally, overall attractiveness was evaluated based on the above results. The Pu in all items of the ADS cycle, regardless of whether they were fresh or spent fuel, had an overall attractiveness of L, which was the same level as the Pu in the spent fuel assembly of BWR-MOX.

### 3. Evaluation of $\sigma_{MUF}$ and its target value

#### 3.1. Methodology

The MBA of the ADS cycle, which was necessary for the evaluation of  $\sigma_{MUF}$ , was designed as shown in **Figure 3**.

Each MBA was separated by either the facility or boundary that switched between bulk and item management. The Pu measurement technologies used in the existing SGs system described in the International Target Values (ITV) provided by the IAEA [21] were installed throughout the MBA. General measurements of nuclear material for the accounting system in SGs are conducted independently by the operator and inspector; therefore, uncertainties accumulate in the results of each measurement. The operators are expected to use the results obtained from the destructive assay (DA) required for fuel quality assurance (green arrows and names in **Figure 3**). Conversely, the inspectors are predicted to actively use a non-destructive assay (NDA) (red arrows and names in **Figure 3**). Thus, the probability density obtained by the inspector is smaller than that obtained by the operator, and the  $\sigma_{MUF}$  for inspectors ( $\sigma_{MUF,I}$ ) is larger than that for operators ( $\sigma_{MUF,O}$ ), as shown in **Figure 4**. The evaluation targets of  $\sigma_{MUF,O}$  and  $\sigma_{MUF,I}$  were MBA-1 and -4, which had bulk management such as powder, pellet, scrap, and salt. The  $\sigma_{MUF,O}$  and  $\sigma_{MUF,I}$  for each MBA were calculated as follows:

$$u_p = m_{Pu,p} \sqrt{\sum_{i=1} \sigma_{p,i}^2} \quad (1)$$

$u_p$ : Absolute uncertainty at measurement point  $p$ ,  
 $\sigma_{p,i}$ : Relative uncertainty of measurement technology  $i$  at measurement point  $p$ ,  
 $m_{Pu,p}$ : Pu mass flow per inspection frequency at measurement point  $p$ .

$$\sigma_{MUF,O(or I)} = \frac{\sqrt{\sum_{p=1} u_p^2}}{M_{Pu}} \quad (2)$$

$\sigma_{MUF,O(or I)}$ : Assumed relative uncertainty of measurement for operators (or inspectors),  
 $M_{Pu}$ : Pu mass flow per inspection frequency in MBA.

Each uncertainty of measurement technology was the measurement accuracy of inspection for Pu in spent fuel cited from ITV [21], because the Pu attractiveness level of the ADS fuel cycle was the same as that of the spent fuel assembly of BWR-MOX, as shown in **Table 4**. Hypothetical measurement technologies for volume (Volume) and density (Density) were assumed to have an accuracy of

Table 3. Material attractiveness evaluation results for nuclear non-proliferation.

| Theft target                    |  |                 | Processing phase   | Utilization phase |             | Overall attractiveness |
|---------------------------------|--|-----------------|--------------------|-------------------|-------------|------------------------|
| Facility                        | Chemical form {theft item}   | Cycle           | Conversion time    | BCM (kg)          | HC (kW/BCM) |                        |
| BWR <sup>(a)</sup>              | MOX {Assembly (Fresh)}   | -               | [M] <sup>(b)</sup> | 13.6 [H]          | 0.14 [H]    | [M]                    |
|                                 | MOX {Assembly (Spent)}   | -               | [L]                | 15.8 [H]          | 0.30 [H]    | [L]                    |
| RFC<br>or<br>ADS <sup>(a)</sup> | (An+RE+Zr)N <sup>(c)</sup> {Mixed nitride powder, Pellet, Pin, Assembly (Fresh)} | 1 <sup>st</sup> | [L]                | 13.9 [H]          | 0.23 [H]    | [L]                    |
|                                 |  | 5 <sup>th</sup> | [L]                | 13.2 [H]          | 3.59 [M]    | [L]                    |
|                                 | (An+RE+Zr)N {Assembly (Spent), Renitride powder}, An-RE-Cd alloy {Cd cathode}    | 1 <sup>st</sup> | [L]                | 13.5 [H]          | 2.03 [M]    | [L]                    |
|                                 |  | 5 <sup>th</sup> | [L]                | 13.3 [H]          | 3.75 [M]    | [L]                    |

<sup>(a)</sup> Cited from previous research [9], <sup>(b)</sup> The number in [ ] indicates the attractiveness level, <sup>(c)</sup> An: actinides (U, Np, Pu, Am, and Cm); RE: Rare-earth elements (Y, La, Ce, Pr, Nd, Pm, Sm, Eu, and Gd).

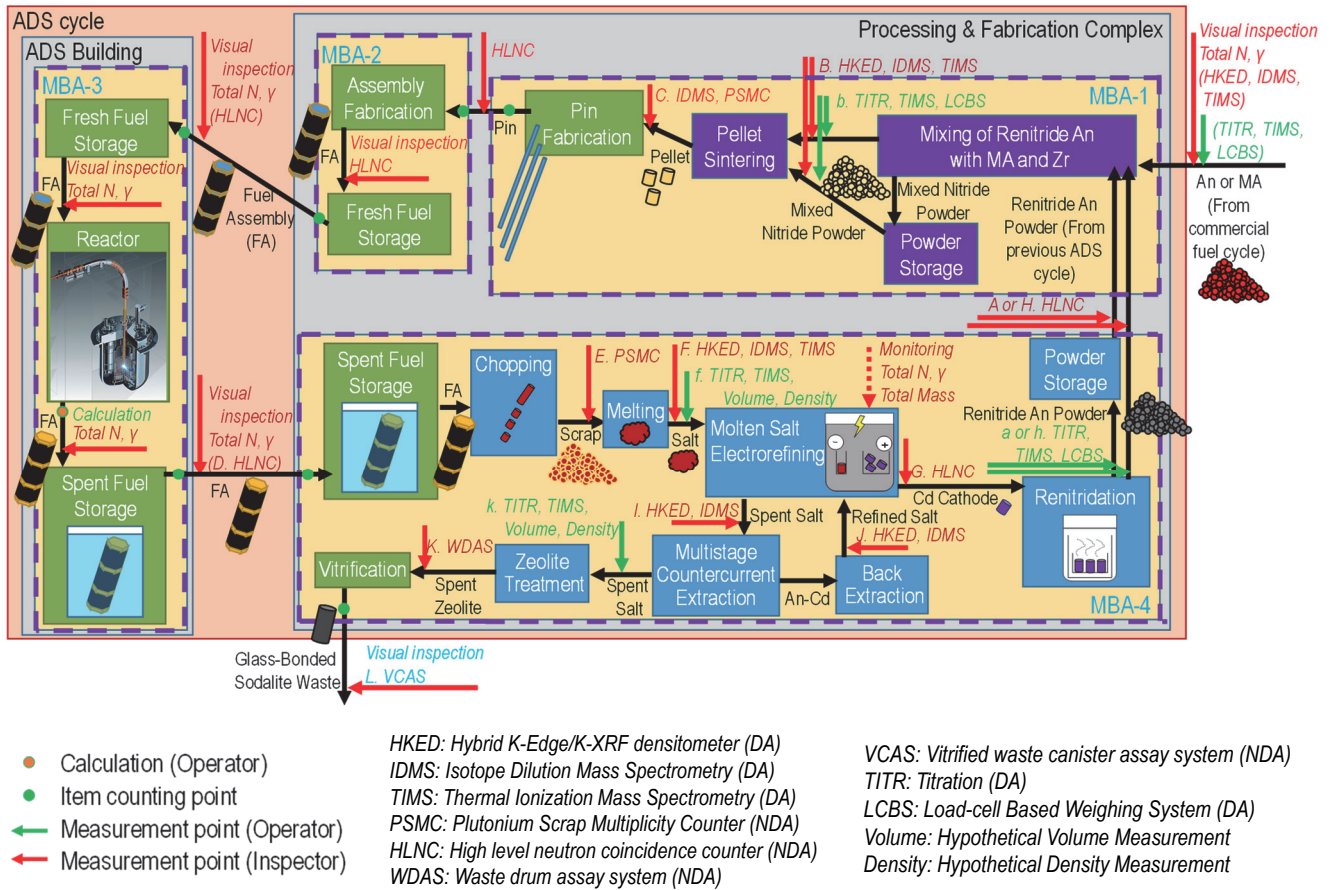


Figure 3. MBA and measurement point for nuclear material inspection.

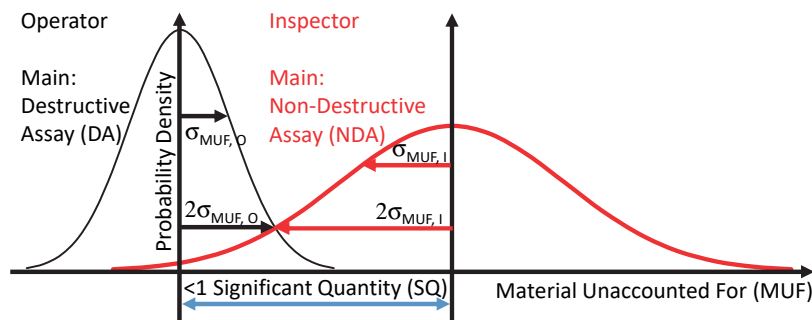


Figure 4. General accuracy requirements for Pu measurement in expected accounting.

1% in this study because there was no suitable technology to measure the volume and density of salt. The Pu mass flow at each measurement point shown in Table 5 was set by assuming each inspection frequency and the latest ADS cycle design.

The expected measurement accuracy requirements for the accounting system in SGs are set such that the sum of  $2\sigma_{\text{MUF},O}$  and  $2\sigma_{\text{MUF},I}$  does not exceed equivalent to 1SQ, as shown in Figure 4. However, owing to the peculiarity of the fuel composition in the ADS cycle, it was unclear whether the conventional method could achieve this required general measurement accuracy. Thus, in this study, the evaluated  $\sigma_{\text{MUF},O}$  and  $\sigma_{\text{MUF},I}$  were individually and directly compared with 1SQ (1SQ standard) in this

study to discuss the  $\sigma_{\text{MUF}}$  target value. The 1SQ standards of MBA-1 and -4 were calculated by dividing the IAEA defined 1SQ of Pu (8kg) by the maximum Pu mass flow of each MBA of each inspection frequency (e.g. 579.2 kg/month of MBA-1, 96.3 kg/5 days of MBA-4, etc.), respectively.

### 3.2. Results

The evaluation results of  $\sigma_{\text{MUF},O}$  and  $\sigma_{\text{MUF},I}$  are shown in Figure 5. The  $\sigma_{\text{MUF},O}$  (MBA-1;2.66%, MBA-4;3.01%) estimated with only the uncertainties of the DAs was more accurate than the  $\sigma_{\text{MUF},I}$  (MBA-1;5.95%, MBA-4;6.78%), which includes the uncertainties of the NDAs. However,

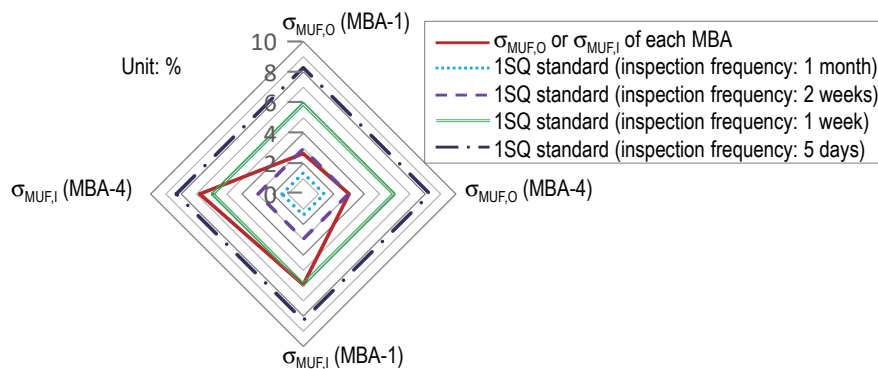


Table 4. Assumptions associated with the measurement method and uncertainty of the inspection technology for nuclear material in ADS cycle [21].

| Measurement method                            | Destructive (DA) or Non-destructive (NDA) assay | Uncertainty (%) |
|---|---|-----------------|
| Hybrid K-edge densitometry (HKED)             | DA  | 0.94            |
| Isotopic dilution mass spectrometry (IDMS)    | DA  | 0.28            |
| Thermal ionization mass spectrometry (TIMS)   | DA  | 1.86            |
| Plutonium scrap multiplicity counter (PSMC)   | NDA   | 5.1             |
| High level neutron coincidence counter (HLNC) | NDA   | 2.2             |
| Waste drum assay system (WDAS)                | NDA   | 11              |
| Vitrified waste canister assay system (VCAS)  | NDA   | 14              |
| Titration (TITR)                              | DA  | 0.28            |
| Load-cell based weighing system (LCBS)        | DA  | 0.07            |
| Hypothetical volume measurement (Volume)      | -   | 1               |
| Hypothetical density measurement (Density)    | -   | 1               |

Table 5. Pu mass flow at each measurement point.

| MBA   | Measurement point | Pu mass flow per inspection frequency |              |           |             |
|-------|-------------------|---------------------------------------|--------------|-----------|-------------|
|       |                   | (kg/month)                            | (kg/2 weeks) | (kg/week) | (kg/5 days) |
| MBA-1 | a, b, A, B, or C  | 579.2                                 | 270.3        | 135.1     | 96.5        |
| MBA-4 | f, D, E, or F     | 577.6                                 | 269.5        | 134.8     | 96.3        |
|       | h, G, H, or I     | 577.5                                 | 269.5        | 134.8     | 96.3        |
|       | J                 | 26.8                                  | 12.5         | 6.3       | 4.5         |
|       | k, K, or L        | 0.1                                   | 0.05         | 0.02      | 0.02        |

Figure 5. Comparison among  $\sigma_{\text{MUF},\text{O}}$  and  $\sigma_{\text{MUF},\text{I}}$  of Pu measurement and their target standard values in equilibrium period.

both  $\sigma_{\text{MUF},\text{O}}$  and  $\sigma_{\text{MUF},\text{I}}$  did not satisfy the 1SQ standard of monthly inspection frequency (MBA-1; 1.38%, MBA-4; 1.39%) because the value of measurement accuracy for spent fuel was conservatively quoted from ITV in anticipation of the difficulty in measuring ADS fuel containing many MAs and REs. Another aspect was that these 1SQ standard values were set too low because the Pu mass flow in a month for each MBA was large. The 1SQ standard values could be relaxed by increasing the inspection frequency, although this would increase the workload of operators and inspectors. As a result, it became clear that the  $\sigma_{\text{MUF},\text{O}}$  satisfied the 1SQ standard by setting the inspection frequency to about two weeks

(MBA-1; 2.96%, MBA-4; 2.97%). Similarly, it was clarified that the inspection frequency should be about 5 days (MBA-1; 8.29%, MBA-4; 8.31%) in order for  $\sigma_{\text{MUF},\text{I}}$  to achieve the 1SQ standard.

#### 4. Conclusions

We evaluated the overall attractiveness assuming the diversion of fuel items from the ADS cycle, considering the fundamental fuel property as an important factor for determining the inspection goal for SGs required for the facility. The Pu in all items of the ADS cycle had an overall attractiveness of L, which is the same level as the Pu in the

spent fuel assembly of MOX for conventional BWR, despite being either fresh or spent fuel. Additionally, the  $\sigma_{\text{MUF},\text{O}}$  and  $\sigma_{\text{MUF},\text{I}}$ , each representing the accumulated uncertainty of measurements by operators and inspectors for the Pu mass flow in the ADS cycle, were hypothetically and conservatively derived with reference to the accuracy targets for existing Pu measurements and the attractiveness evaluation results to design and evaluate accounting systems for SGs.  $\sigma_{\text{MUF},\text{O}}$  was more accurate than  $\sigma_{\text{MUF},\text{I}}$  because NDAs conducted only by inspectors had relatively large uncertainties. We also discussed the target values for both  $\sigma_{\text{MUF},\text{O}}$  and  $\sigma_{\text{MUF},\text{I}}$  using the 1SQ standard values to estimate the acceptable inspection frequency. As a result, it was found that the  $\sigma_{\text{MUF},\text{O}}$  and  $\sigma_{\text{MUF},\text{I}}$  satisfied each 1SQ standard by setting the inspection frequencies to about 2 weeks and 5 days, respectively.

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