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ARTICLE

Development of a method for the determination of spontaneous fission nuclides in irradiated fuel and applicability to Pu quantification in fuel debris by dual time neutron measurements

Taketeru Nagatani [a, b], Yoshihiro Kosuge [c], Hiroshi Sagara [a *], Sho Nakaguki [b], Takayoshi Nohmi [b] and Keisuke Okumura [b]

[a] *Tokyo Institute of Technology, 2-12-1-N1-17 Ookayama, Meguro-ku, Tokyo 152-8550, Japan;* [b] *Japan Atomic Energy Agency, 4-33 Muramatsu, Tokai-mura, Naka-gun, Ibaraki 319-1194, Japan;* [c] *NESI, 38 Shinkocho Hitachinaka-shi, Ibaraki 312-0005, Japan*

This study provides an overview of plutonium quantification in irradiated fuel, including fuel debris, at the Fukushima Daiichi Nuclear Power Plants, named the Differential Half-life of Spontaneous Fission Nuclides method. Spontaneous fission nuclides in irradiated fuel decrease exponentially with time. Using the difference in half-life of each nuclide, Pu-240 effective mass can be quantified by two neutron measurements with long time intervals. The Pu mass can be quantified by utilizing the correlation between the mass ratio of Cm-244/ Pu-240 effective and the mass ratio of Pu/ Pu-240 effective. The applicability of the Differential Half-life of Spontaneous Fission Nuclides method is numerically evaluated. The results show that a long-time interval is required to reduce random errors. When the interval between the first and second measurements is 32 years, the Pu-240 effective mass and Pu mass can be quantified with uncertainties of 10-50% depending on the presence of water in the storage canister and the burnup condition of the irradiated fuel, including a mixture of several burnup compositions in fuel debris.

Keywords: severe accident; Nuclear Power Plant; MCNP; Fukushima Daiichi Nuclear Power Plants; fuel debris; passive neutron; non-destructive assay; differential half-life of spontaneous fission nuclides method

1. Introduction

1.1. Needs for quantification of plutonium in fuel debris

To decommission the Fukushima Daiichi Nuclear Power Plants (1F), measurement technology to quantify the nuclear material in fuel debris will be required for appropriate nuclear material management. It is challenging to quantify nuclear material in fuel debris using conventional destructive assay (DA) and non-destructive assay (NDA) methods. Fuel debris is considered solidified and mixed with surrounding materials, such as zircaloy and concrete [1]. Therefore, it is considered insoluble and inhomogeneous, making it unsuitable to apply DA for fuel debris [2]. Moreover, fuel debris contains minor actinides and fission products, which are intense neutron and gamma ray sources generated by fuel burnup in a reactor. It also contains neutron absorbers, such as B-10, in the control rods. These materials make it difficult to apply conventional NDA to fuel debris. It is difficult to accurately quantify the nuclear material in fuel debris by applying a single measurement technology. Therefore, an integrated measurement system is required to combine passive neutrons, active neutrons, passive gamma, and active gamma techniques. To develop an integrated measurement system, it is important to determine characteristics such as the range of application and uncertainty for each technology. We conducted a characterization study for each technology based on a numerical evaluation [3]. In this study, we numerically evaluated the applicability of the passive neutron technique to quantify plutonium in fuel debris.

1.2. Challenges to quantification of plutonium in fuel debris

From the viewpoint of NDA based on neutron measurements, it is challenging to quantify plutonium because of the following two issues.

- The amount of fissile material, neutron poison, and water in each canister varies; thus, the neutron leakage multiplication varies.
- The ratio of spontaneous fission neutrons of Cm-244, the dominant neutron source, to that of the Pu-240 effective (*Pu-240e*) increased exponentially as burnup increased as shown in **Figure 1**. When multiple burnups are mixed, the ratio does not correlate with burnup. Equation 1 defines *Pu-240e* mass as follows:

*Corresponding author. E-mail: sagara.h.aa@m.titech.ac.jp

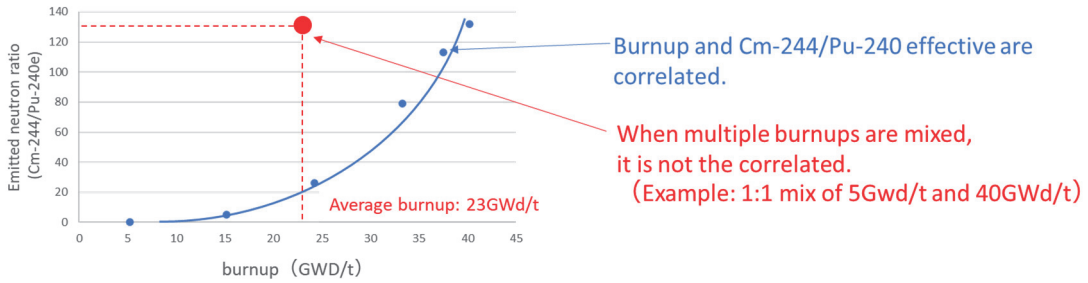

Figure 1. Correlation between neutron emission ratio of Cm-244 to *Pu-240e* mass and burnup.

$$Pu\text{-}240e\ mass = \left(\frac{SFY_{^{238}Pu}}{SFY_{^{240}Pu}} \cdot f_{^{238}Pu} + \frac{SFY_{^{240}Pu}}{SFY_{^{240}Pu}} \cdot f_{^{240}Pu} + \frac{SFY_{^{242}Pu}}{SFY_{^{240}Pu}} \cdot f_{^{242}Pu}\right) \cdot m_{total-Pu} \quad (1)$$

Pu-240e mass: Pu-240 effective mass (g)
SFY: Spontaneous fission yield (n/s-g); Pu-238: 2.59×10^3, Pu-240: 1.02×10^3, Pu-242: 1.72×10^3 [4]
f: Weight fraction of nuclides to total mass of Pu
$m_{total\text{-}Pu}$: Total mass of Pu (g)

1.3. Scope of previous studies and this study

Figure 2 shows the overall concept of plutonium quantification. In previous studies, uncertainty of quantification of spontaneous fission nuclides (Cm-244 effective: *Cm-244e*) mass was evaluated. Equation 2 defines *Cm-244e* mass. The leakage multiplication varies depending on the shape and composition of the fuel debris. The applicability of Differential Die-Away Self-Interrogation (DDSI) technique for determining leakage multiplication and the uncertainty for quantification of *Cm-244e* mass was evaluated [5].The purpose of this study is to evaluate a method for quantification of *Pu-240e* mass and plutonium mass.

$$Cm\text{-}244e\ mass = \left(\frac{SFY_{^{238}Pu}}{SFY_{^{244}Cm}} \cdot f_{^{238}Pu} + \frac{SFY_{^{240}Pu}}{SFY_{^{244}Cm}} \cdot f_{^{240}Pu} + \frac{SFY_{^{242}Pu}}{SFY_{^{244}Cm}} \cdot f_{^{242}Pu} + \frac{SFY_{^{244}Cm}}{SFY_{^{244}Cm}} \cdot f_{^{244}Cm}\right) \cdot m_{total\text{-}Pu\ and\ Cm} \quad (2)$$

Cm-244e mass: Cm-244 effective mass (g)
SFY: Spontaneous fission yield (n/s-g); Cm-244: 1.08×10^7 [4]
f: Weight fraction of nuclides to total mass of Pu and Cm
$m_{total\text{-}Pu\ and\ Cm}$: Total mass of Pu and Cm(g)

2. Development of methodology of plutonium quantification

2.1. Quantification of Pu-240e mass

Cm-244e mass in the irradiated fuel debris decreased with time, according to an exponential function. The half-lives of the main component nuclides in *Cm-244e* are shown below [6].

- Cm-244: approx.18.11 years ±0.03
- Pu-238: approx.87.7 years±0.1

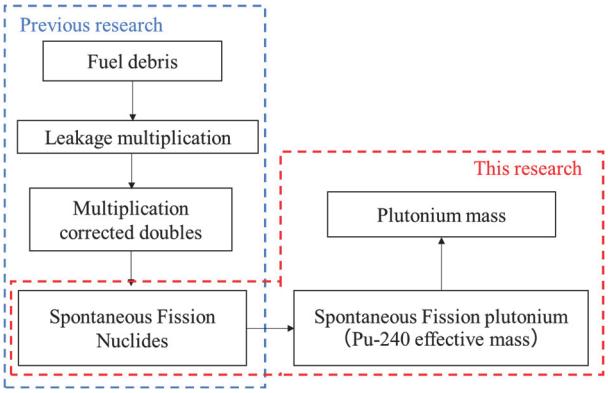

Figure 2. Overall concept of plutonium quantitation.

- Pu-240: approx.6,561 years±7
- Pu-242: approx.3.735×10^5 years±0.011

It is assumed that the decrease in *Cm-244e* mass is due to the decrease in Cm-244, as Cm-244 has a much shorter half-life than Pu-238, 240, 242. By measuring *Cm-244e* mass twice within a time interval, the exponential function for attenuation of Cm-244 can be estimated, and *Pu-240e* mass and Cm-244 mass can be quantified. This method is named Differential Half-life of Spontaneous Fission Nuclides (DHS) method. **Figure 3** shows a conceptual diagram of the DHS method.

2.2. Quantification of Pu mass

As shown in Figure 1, the mass ratio of Cm-244 to *Pu-240e* (*Cm-244/Pu-240e*) was correlated with burnup. The mass ratio of Pu to *Pu-240e* (*Pu/Pu-240e*) was also correlated with burnup. *Cm-244/Pu-240e* and *Pu/Pu-240e* were correlated via burnup. **Figure 4** shows the correlation between *Cm-244/Pu-240e* and *Pu/Pu-240e*. Pu mass can be quantified by this correlation and mass of Cm-244 and *Pu-240e* obtained by the DHS method.

3. Evaluation method

In this study, uncertainties of quantification of *Pu-240e* mass and Pu mass were evaluated using the calculated compositions of spontaneous fission nuclides. It should be noted that neutron transport calculations were not performed in this study because uncertainty for quantification of *Cm-244e* mass have been clarified in previous studies [5]. The

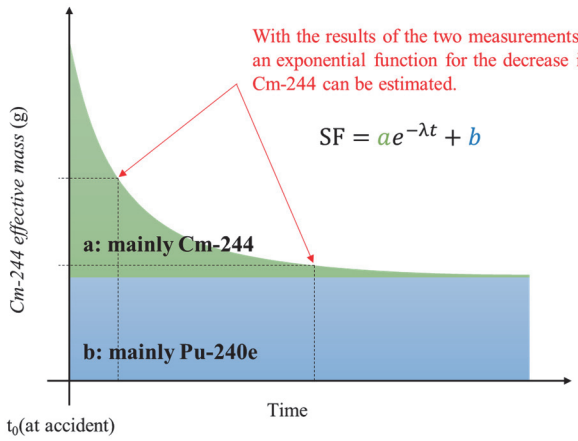

Figure 3. Conceptual diagram of DHS method.

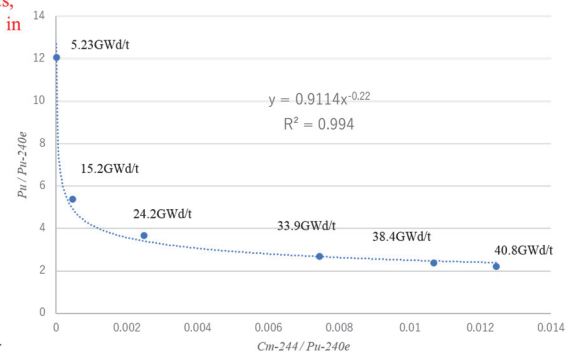

Figure 4. Correlation between Cm-244/*Pu-240e* and Pu/*Pu-240e*.

evaluation steps were as follows:

A) The compositions of spontaneous fission nuclides were calculated ORIGEN 2.2 under the following conditions.
- Initial enrichment: 3.7%
- Burnup: Six different burnups of unit 1 of 1F, which is an equilibrium core of 5.23, 15.2, 24.2, 33.9, 38.4, and 40.8(GWD/t)[7]. Mixture of multiple burnups were also evaluated for the following cases.
  - Mixed case 1: mixture of six different burnup compositions with the same amount (average burnup: 26.3GWd/t)
  - Mixed case 2: mixture of lowest burnup compositions and highest burnup compositions in a ratio of 16:1 (average burnup: 7.32GWd/t)
- Time frame: It was assumed that the first measurement was 10 years after the accident and the second was Δt years after the first. Δt are 2, 4, 8, 16, and 32 years.
- Target spontaneous fission nuclides: U-232, U-233, U-234, U-235, U-236, U-238, Np-237, Pu-236, Pu-238, Pu-239, Pu-240, Pu-241, Pu-242, Pu-246, Am-241, Cm-242, Cm-244, Cm-246, Cm-248, Cf-250, Cf-252 and Cf-254.

B) Taking into account the Spontaneous fission yield of each nuclide, *Cm-244e* mass, composed of all target spontaneous fission nuclides, was calculated from the spontaneous fission nuclide mass.

C) "*a*" and "*b*" in Figure 3 was evaluated from the simultaneous equations by substituting t_1 and *Cm-244e mass*$_1$, t_2 and *Cm-244e mass*$_2$, and λ:1.213e-09 (decay constant of Cm-244) into Eq. (3).

$$Cm-244e\ mass = ae^{-\lambda t} + b \quad (3)$$

D) *Pu-240e* mass was evaluated by Equation 4.

$$Pu\text{-}240e\ mass = b \times SFY_{^{244}Cm} \div SFY_{^{240}Pu} \quad (4)$$

E) Pu mass was evaluated by substituting *Pu-240e* mass and Cm-244 mass, as evaluated by the DHS method, into Equation 5. Cm-244 mass was evaluated from "*a*" by Eq. (3).

$$Pu\ mass = 0.9114 \times \left({}^{Cm-244}/_{Pu-240_e}\right)^{-0.22} \times Pu-240_e \quad (5)$$

F) Systematic errors for *Pu-240e* mass and Pu mass were evaluated from relative difference between the evaluated value and true value.

G) Random errors for *Pu-240e* mass and Pu mass were evaluated from relative standard deviation. The σ(*Cm-244e* mass) was assumed to have a counting error of 0.3% for wet storage with canisters filled with water or 0.04% for dry storage with canisters filled with air for 20-minute measurements evaluated in previous studies [5].

H) Total measurement uncertainty (TMU) was evaluated by the square root of the sum of the squares of systematic errors and random errors.

4. Evaluation result

4.1. Uncertainty of quantification of Pu-240e mass

Figure 5 shows the uncertainty of quantification of *Pu-240e* mass for wet storage. Random error improved with increasing Δt. A longer time interval resulted in a more significant difference in *Cm-244e* mass between the first and second measurements. This larger difference improved the prediction accuracy of attenuation formula. Furthermore, random error degraded with increasing burnup. This is because *Pu-240e*/Cm-244 ratio, in other words *b/a* ratio in Figure 3, decreases with increasing burnup.

For the systematic error, *Pu-240e* mass was overestimated with increasing burnup. **Figure 6** shows the correlation between the relative difference for *Pu-240e* mass and burnup and the correlation between neutron emission ratio of Cm-246 to *Pu-240e* and burnup. The ratio of Cm-246 to *Pu-240e* increases with increasing burnup. Because Cm-246 has a long half-life, approximately 4,760 years ±40 [6], it was included in the "*b*" in Fig. 3, and *Pu-240e* mass was overestimated.

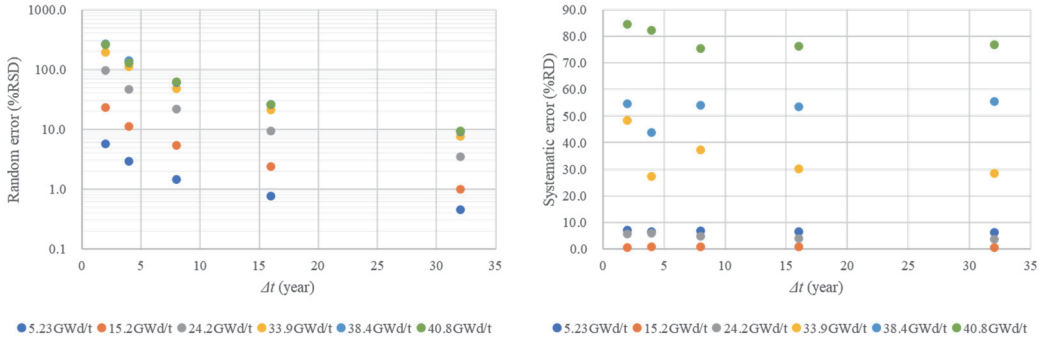

Figure 5. Uncertainty of quantification of *Pu-240e* mass for wet storage (left: random error, right: systematic error).

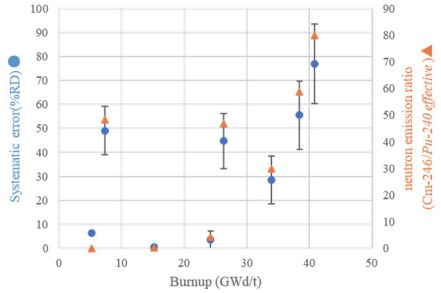

Figure 6. Relationship between systematic error and neutron from Cm-246 (Δt=32 year).

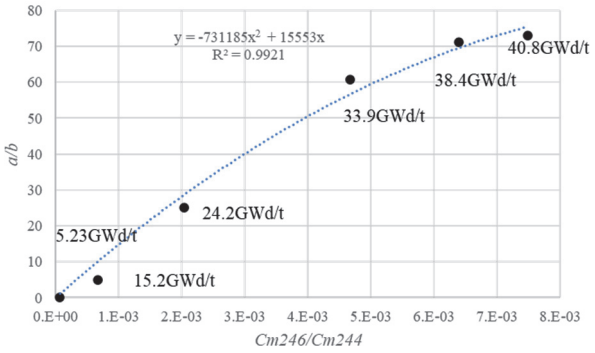

Figure 7. Correlation between *a/b* and *Cm-246/Cm-244*.

4.2. *Development of correction method of impact of Cm-246 mass on Pu-240e mass*

Figure 7 shows the correlation between *a/b* in Figure 3 and the mass ratio of Cm-246 to Cm-244 (*Cm-246/Cm-244*). Using this correlation, the Cm-246 mass was evaluated and *Pu-240e* mass was corrected. **Figure 8** shows the systematic error for corrected *Pu-240e* mass. The correction significantly improved the systematic error. However, for mixed case 2, the systematic error was still larger compared with others because the correction used the correlation with burnup.

4.3. *Uncertainty of quantification of Pu mass*

Figure 9 shows the systematic error for quantification of Pu mass for wet storage. As well as the systematic error for corrected *Pu-240e mass*, the mixed case 2 has larger systematic error compared with others due to the large relative difference on corrected *Pu-240e* mass.

4.4. *TMU of quantification of Pu-240e mass and Pu mass*

Tables 1 and **2** show uncertainties of quantification of *Pu-240e* mass and Pu mass in typical cases for wet and dry storage, respectively. TMU of quantification of *Pu-240e* mass was better than that of Pu mass due to more direct measurement and simpler evaluation process. For the low-burnup, random errors were minor, and systematic errors were dominant. For high-burnup, systematic errors were equivalent to low burnup, and random errors were dominant. Therefore, a long-time interval is required to reduce random errors. The random errors for dry storage

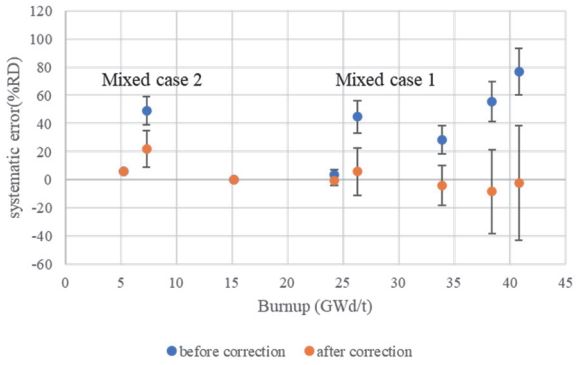

Figure 8. Systematic error for corrected *Pu-240e* mass (Δt=32 year).

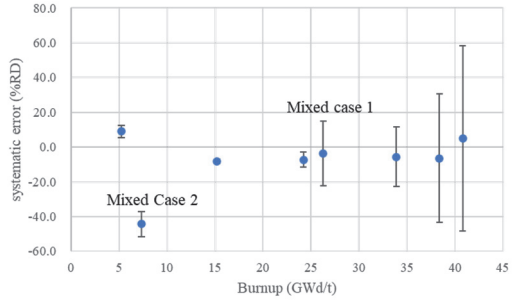

Figure 9. Systematic error for quantification of *Pu* (Δt=32 year).

Table 1. Uncertainties of quantification of *Pu-240e* mass and Pu mass in typical cases for wet storage.

Burnup (GWd/t)	Δt (year)	*Pu-240e* mass			Pu mass		
		Random error (%RSD)	Systematic error (%RD)	TMU (%)	Random error (%RSD)	Systematic error (%RD)	TMU (%)
Low burnup (5.23GWD/t)	8	1.5	6.7	6.9	9.5	11.3	14.8
	32	0.5	6.3	6.3	3.1	9.0	9.5
High burnup (40.8GWD/t)	8	226.8	-3.9	226.8	277.1	2.8	277.1
	32	32.8	-0.7	32.8	40.1	7.0	40.7
Mixed case 1	32	16.0	4.8	16.7	19.6	-5.0	20.2
Mixed case 2	32	10.7	21.2	23.7	13.2	-44.8	46.7

Table 2. Uncertainties of quantification of *Pu-240e* mass and Pu mass in typical cases for dry storage.

Burnup (GWd/t)	Δt (year)	*Pu-240e* mass			Pu mass		
		Random error (%RSD)	Systematic error (%RD)	TMU (%)	Random error (%RSD)	Systematic error (%RD)	TMU (%)
Low burnup (5.23GWD/t)	8	0.2	6.7	6.7	1.3	11.3	11.4
	32	0.1	6.3	6.3	0.4	9.0	9.0
High burnup (40.8GWD/t)	8	30.2	-3.9	30.5	36.9	2.8	37.1
	32	4.4	-0.7	4.4	5.4	7.0	8.8
Mixed case 1	32	2.1	4.8	5.2	2.6	-5.0	5.6
Mixed case 2	32	1.4	21.2	21.2	1.8	-44.8	44.8

were much smaller than those for wet storage because of the small counting error of dry storage. This indicated that dry storage is advantageous for the application of the DHS method. For single burnup, TMU for *Pu-240e* mass and Pu mass was small enough. This indicated that DHS method has a capability to quantify Pu mass for spent fuel with low uncertainty.

In order to evaluate the applicability of DHS method to fuel debris, it is necessary to evaluate the case with a mixture of multiple burnups. For mixture of multiple burnups, systematic errors were degraded. Especially, large systematic errors exist in mixtures containing a large amount of low burnup compositions and a small amount of high burnup compositions. This indicated that there is possibility that uncertainty of DHS method for fuel debris may be degraded up to 50%.

5. Conclusion

Focusing on the difference in the half-life of spontaneous fission nuclides, we developed the DHS method to quantify mass of *Pu-240e* and Pu by evaluating *Cm-244e* mass twice with time intervals by neutron measurement. The results of the applicability evaluation revealed that the DHS method has capability to quantify mass of *Pu-240e* and Pu in irradiated fuels, including fuel debris. The quantification uncertainty mainly depended on the length of the time interval between the two measurements, the presence of water in the storage canister, and the burnup condition of the irradiated fuel. The dry storage is advantageous for the application of the DHS method due to the low random error. Mixture of multiple burnups degraded the systematic error. When the time interval between the first and second measurements is 32 years, *Pu-240e* and Pu can be quantified with uncertainties of 10-50% depending on the presence of water in the storage canister and the burnup condition of the irradiated fuel, including a mixture of multiple burnup compositions in fuel debris.

This evaluation assumes that *Cm-244e* mass can be accurately quantified. However, it should be noted that there are factors that increase systematic errors that are not considered in this evaluation, such as changes in neutron leakage multiplication owing to changes in the shape of fuel debris in the storage canister and changes in the environment surrounding the measurement when the time interval is long.

This method cannot quantify Pu at the time of removal. However, it is expected to contribute to improving the accuracy and reliability of material accountancy data during long-term storage because fuel debris is expected to be stored for a long time after removal until treatment and disposal.

References

[1] TEPCO, [Online] Available: https://www.tepco.co.jp/en/hd/decommission/progress/retrieval/index-e.html.

[2] Task Force on Research Strategy for Debris of Fukushima Daiichi Nuclear Power Station, Analysis of Debris Samples of Tokyo Electric Power Company Holdings Fukushima Daiichi Nuclear Power Station, *JAEA-Review 2020-055* (2020), Japan Atomic Energy Agency.

[3] T. Nagatani, M. Komeda, T. Shiba, Y. Nauchi, M. Maeda, H. Sagara, Y. Kosuge, M. Kureta, H. Tomikawa, K.

Okumura and C. Heinberg, Characterization Study of Four Candidate Technologies for Nuclear Material Quantification in Fuel Debris at Fukushima Daiichi Nuclear Power Station, *Energy Procedia* 131 (2017), pp. 258-263.

[4] N. Ensslin, W.C. Harker, M.S. Krick, D.G. Langner, M.M. Pickrell and J.E. Stewart, Application Guide to Neutron Multiplicity Counting, LA-13422-M, (1998), Los Alamos National Laboratory.

[5] T. Nagatani, H. Sagara, Y. Kosuge, T. Nohmi and K. Okumura, Applicability of Differential Die-Away Self-Interrogation Technique for Quantification of Spontaneous Fission Nuclides for Fuel Debris at Fukushima Daiichi Nuclear Power Plants, *Journal of Nuclear Science and Technolgy* 60 (2023), pp. 460-472.

[6] M. Namekawa and T. Fukahori, Tables of Nuclear Data (JENDL/TND-2012), JAEA-Data/Code 2012-014, (2012), Japan Atomic Energy Agency.

[7] K. Nishihara, H. Iwamoto and K. Suyama, Estimation of fuel compositions in Fukushima-Daiichi Nuclear Power Plant, JAEA-Data/Code 2012-018, (2012).