I-1. PROJECT RESEARCHES

Project 6

PR6

Fundamental Research on Decommissioning of Reactor Facility

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OBJECTIVES and RESEARCH SUBJECTS:

The main objective of this research project is to improve the method and the fundamental information on decommissioning of KUR and the Fukushima-Daiichi NPPs. This project consists of seven research subjects from four institutions, as follows.

R5P6-1: Neutron Flux Measurement for Activation Modeling of KUR facility (T. Sano *et al*., Kindai Univ.)
R5P6-2: Measurement of Activation in the Structure Materials of Reactor for decommissioning Process (K. Takamiya *et al*., Kyoto Univ.)
R5P6-3: Fundamental Research about Radioactive Contamination Survey on Nuclear Reactor Facility (S. Fukutani *et al*., Kyoto Univ.)
R5P6-4: Neutron Capture Cross-Section Measurements with TC-Pn in KUR for Same Nuclides targeted for Decommissioning (S. Nakamura *et al*., JAEA)
R5P6-5: Identification of Nuclear Material in Spent Fuel (Y. Nauchi *et al*., Central Research Institute of Electric Power Industry)
R5P6-6: External Neutron Source for Non-destructive Analysis of Fuel Debris (Y. Nauchi *et al*., Central Research Institute of Electric Power Industry)
R5P6-7: Study on Non-destructive Analysis Method for Fuel Debris using Neutron Resonance Absorption (J. Hori *et al*., Kyoto Univ.)

One research project (R5P6-2) could not be reported in this fiscal year since a sampling inspection of the object is under way.

MAIN RESULTS and CONTENTS of this REPORT:

T. Sano *et al.* (R5P6-1) evaluated the neutron flux distribution in the KUR sub-pile room during 5-MW operation for the decommissioning of KUR.
S. Fukutani *et al.* (R5P6-3) measured stable nuclide by INAA (Instrumental neutron activation analysis) for decommissioning the KUR stack made of RC (Reinforced Concrete). About 100 mg core samples were irradiated with standard samples by using Pn-2 and Pn-3 of KUR.
S. Nakamura *et al.* (R5P6-4) performed the activation experiments by using the thermal column pneumatic tube (TC-Pn) of KUR. The tentative thermal-neutron capture cross-sections of ^{58}Fe, ^{170}Er and ^{180}Hf selected from the viewpoint of clearance level in decommissioning were obtained.
Y. Nauchi *et al.* (R5P6-5) measured the gamma-ray spectrum from short-lived FPs from fission of ^{235}U induced by thermal neutrons at the pulsed neutron source in the KURNS-LINAC. The out-of-phase event spectroscope (OOPS) newly developed was applied.
Y. Nauchi *et al.* (R5P6-6) measured the background gamma rays were measured with good energy resolution using a fusion neutron source in an inertial electrostatic confinement device (IEC source) as an external source to apply a neutron induced gamma ray spectroscopy (NIGS) to non-destructive quantification technique of ^{235}U enrichment of irradiated nuclear material.
J. Hori *et al.* (R5P6-7) performed the verification experiment with a self-indication method for applying to the non-destructive assay of nuclear material such as fuel debris using a pulsed neutron source in the KURNS-LINAC. It was confirmed that the estimation of the reduction rate can be used for the determination of the areal density of fuel material.

PR6-1

Neutron flux measurement for KUR facility activation modeling

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INTRODUCTION: For the decommissioning of the KUR, it is important to evaluate the amount of radioactivation in the generated waste. In particular, it is important to evaluate the radioactivation in the equipments around the biological shield and the neutron tubes. In general, the evaluation of the activation dose is mainly based on numerical calculations (deterministic or probabilistic models), although some part of the activation dose is measured. In numerical calculations, the neutron spectrum and absolute neutron flux are important in-put information. In recent years, the performance of computers has improved to the point where it is now possible to calculate neutron flux for the entire facility, including buildings, and to evaluate the injected neutron spectrum into facilities and equipment. However, the absolute values often differ by several 10% or more between the calculated and measured values. In the 2022, the neutron flux on the KUR biological shielding surface was measured using the EneRgY selective Neutron detector containment device for thermal Group neutron InterruptIon "ERYNGII" neutron detector [1].
In 2023, neutron shielding calculations were performed using Monte Carlo method to predict the neutron flux in the sub-pile room under the KUR.

CALCULATIONS: Neutron flux calculations in the KUR subpile room were performed in the following two steps: 1) A core calculation of KUR was performed using the Monte Carlo calculation code MVP3 [2] and JENDL-5 [3] to obtain the neutron spectrum in the KUR core. 2) Using the neutron spectrum as input, a neutron transport calculation was performed using PHITS2 version 3.31 [4] and JENDL-5 to obtain the neutron flux in the KUR sub-pile room.
Here, the core conditions of the KUR were adopted for the core in 2023 year. In addition, for simplification of the calculation model, each irradiation facilities and neutron tubes were not taken into account in the calculations.

RESULTS: Figure 1 shows the neutron flux distribution in the KUR calculated by PHITS2. The value of neutron flux is relative. Neutron flux at the top of the reactor are well shielded by the cooling water. On the other hand, the neutron flux in the sub-pile room during 5 MW operation was evaluated to be about 100 n/cm/sec.

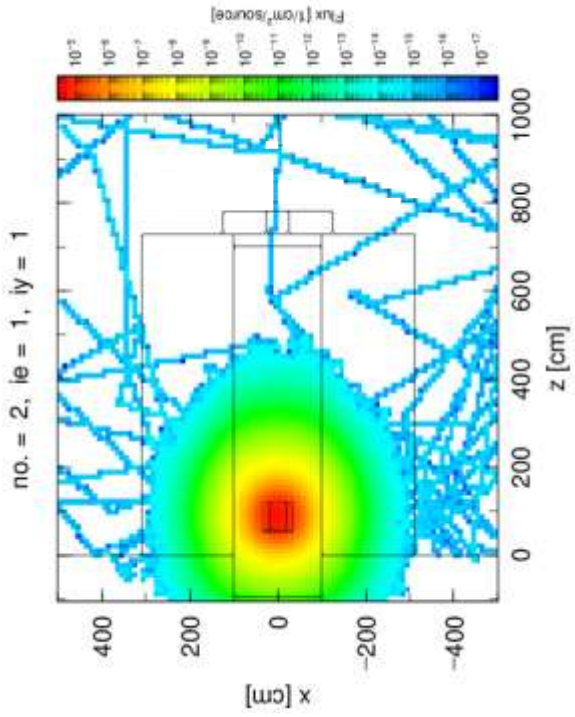

Fig. 1. Calculated neutron flux distribution

REFERENCES:
[1] T. Sano, *et al.,* KURNS-Progress report 2022 (2023) 71.
[2] Y. Nagaya, *et al.*, JAEA-Data/Code 2016-018 (2016).
[3] O. Iwamoto, *et al.,* J. Nucl. Sci. Technol., **60** (2023) 1-60.
[4] T. Sato, *et al.,* J. Nucl. Sci. Technol., **55** (2018) 684-690.

Fundamental Research about Radioactive Contamination Survey on Nuclear Reactor Facility

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INTRODUCTION: It is important to survey radioactive contamination for decommission of nuclear reactor facilities. The KUR stack, which was built in 1963, made of RC (Reinforced Concrete) was demolished in 2013. In this study, we used the KUR stack core [1] and measured stable nuclide by INAA for fundamental estimate of activation amount in concrete.

EXPERIMENTS: Instrumental neutron activation analysis (INAA) was conducted for KUR stack core samples, which was made of RC (Reinforced Concrete). Pn-2 and Pn-3 of KUR facilities were used. For the experiment by using Pn-2, about 100mg core samples were irradiated with standard samples, which were BCR-176 (from Commission of the European Communities, city waste incineration ash) and JB-2 (from National Institute of Advanced Industrial Science and Technology, Basalt). At 5MW thermal power of KUR, samples were irradiated for 60 minutes. After about 6 days and 40 days cooling time, γ spectrometry was made by the HPGe semiconductor detector (ORTEC GEM25185). The experiment by using Pn-3 was conducted for measuring chlorine (Cl), and Cl-38 was measured. Standard sample for Cl was used LiCl solution (1003mg-Li / L). At 1MW thermal power of KUR, samples were irradiated for 30 seconds. Cl-38 is short half-life nuclide (half-life is about 37.2 minutes), so after irradiation, the measurement by γ spectrometry with the HPGe semiconductor detector (ORTEC GEM25185) was made Immediately. Quantitative of each element was conducted by comparative method

RESULTS: Main measurement results by INAA ware shown in the Table 1. Arsenic (As) is short half-life nuclide, so it may be not important as radioactive nuclide. But as element, because of its toxicity, it is necessary to be careful. As for chlorine, Cl-38 is made by neutron activation from stable nuclide Cl-37, abundance: 24.24%, (n, γ) cross section: 0.433b. Cl-38 is very short half-life nuclide, it is not important for radioactive waste treatment and disposal. But, chlorine has another stable nuclide Cl-35. Abundance and (n, γ) cross section of Cl-35 are 75.76% and 43.6b. Cl-36 made from Cl-35 is long half-life (3×10^5 year), pure beta emitter nuclide. It is difficult to detect and measure. Cl-38 wad detected by neutron activation, and it means Cl-35 was made at the same time. And chlorine is thought to be negative ion form and to be easy mobility, it is necessary to be careful.

Table 1 Measuring Results by INAA

Element	As	Ce	Cr	Co	Eu
Nuclide	As-76	Ce-144	Cr-51	Co-60	Eu-152
Half-life	1.1d	285d	28d	5.3y	13y
Conc. (mg/kg)	8.38 ± 1.7	6.80 ± 1.07	5.56E+1 ± 4.0	8.75 ± 7.6E-2	0.44 ± 3.4E-2
Element	Mn	Sb	Zn	Cl	
Nuclide	Mn-54	Sb-124	Zn-65	Cl-38	
Half-life	312d	60d	244d	37m	
Conc. (mg/kg)	7.17E+2 ± 7.3E+1	5.69 ± 0.15	3.40E+2 ± 4.8	2.34E+2 ± 1.1E+2	

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[1] S. Fukutani *et al.,* KURNS Prog. Report 2022 (2023) 72.

Neutron Capture Cross-Section Measurements at TC-Pn in KUR for Nuclides of Concern in Decommissioning

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INTRODUCTION: As nuclear facilities are dismantled in decommissioning, of course, various and large amounts of waste are generated, for instance, structural materials, concrete pieces, pipes, electric cables and so on. Even more inconveniently, they are radioactive waste due to neutron activation. In decommissioning, it is necessary to predict and evaluate the amount of radioactivity generated. Thus, the present study aims to measure accurate neutron capture cross-sections for nuclides of concern in decommissioning. We have performed measurements using the thermal column pneumatic tube (TC-Pn) of KUR [1, 2]. From the viewpoint of clearance level [3] in decommissioning, the present work selected the following nuclides and measured their thermal-neutron capture cross-sections: ^{58}Fe, ^{170}Er and ^{180}Hf nuclides.

EXPERIMENTS: High purity metal samples were prepared for irradiation. A gold-aluminum alloy wire and a cobalt foil were used to monitor the neutron flux at the irradiation position. **Fig. 1** draws a rough sketch of irradiation targets and TC-Pn. Two targets were prepared, (a) and (b). To make use of the well-thermalized neutron field, two dummy capsules were sent into TC-Pn in advance, and then followed by the one target capsule. The target was irradiated for 1 hour under 1-MW operation of KUR. The other target capsule was irradiated in the same condition next day. After irradiation, the samples and flux monitors were enclosed in a thin vinyl bag one by one. The sample was subjected to γ-ray measurement with a high-purity Ge detector. A sample was placed at a distance of 100 mm from the front surface of the Ge detector. The γ-ray peak efficiencies of the Ge detector were measured with a gamma reference source of ^{152}Eu and a mixed γ-ray source.

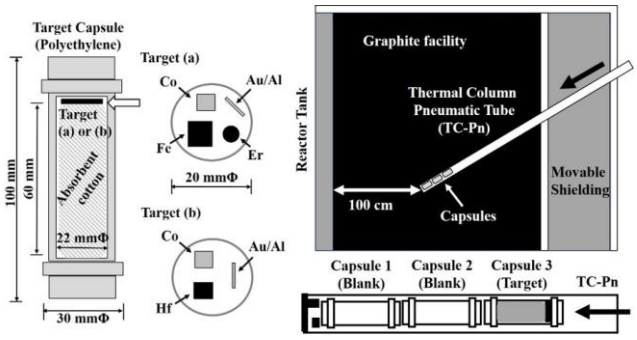

Fig.1 Rough sketch of targets and TC-Pn facility.

ANALYSIS AND RESULTS: The reaction rates of the samples and flux monitors were obtained from their γ-ray yields. Using the reaction rates of monitors, the thermal-neutron flux component was derived on the basis of *Westcott*'s convention [4]. The obtained rection rates for samples divided by the thermal-neutron flux component present the thermal-neutron capture cross-sections. Details of the analysis are given in Ref. [1]. The tentative results were obtained as follows: 1.23±0.03 barn for the ^{58}Fe(n,γ)^{59}Fe reaction, 8.19±0.35 barn for the ^{170}Er(n,γ)^{171}Er reaction and 13.57±0.14 barn for the ^{180}Hf(n,γ)^{181}Hf reaction. As a by-product, 0.427±0.006 barn for the ^{179}Hf(n,γ)^{180m}Hf reaction was obtained. The authors currently proceed with the analysis toward derivation of the final results of the thermal-neutron capture cross-sections for those reactions.

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[1] S. Nakamura *et al*., J. Nucl. Sci. Technol., **58(10)** (2021) 1061.
[2] S. Nakamura *et al*., J. Nucl. Sci. Technol., **59(11)** (2022) 1388.
[3] IAEA; 2004. (No. RS-G-1.7).
[4] C.H. Westcott *et al.,* "*Proc.2nd Int. Conf. Peaceful Use of Atomic Energy, Geneva*", **16** (1958) 70.

Identification of Nuclear Material in Spent Fuel (2)

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INTRODUCTION: The delayed gamma ray assay would be useful to estimate fission rate ratio of nuclides in nuclear fuel. For the assay, nuclear data of yield and decay of fission products (FPs) are essential. To evaluate those with better accuracy, we have measured γ rays from short-lived FPs radiated from fission of ^{235}U induced by thermal neutrons at the pulsed source in the KURNS-LINAC facility [1]. For that, the **o**ut-**o**f-**p**hase event **s**pectroscopy (OOPS) has been newly developed where γ ray spectra are measured after thermal neutron decays out. In the present work, pulse frequency F is surveyed to enhance the counting efficiency.

EXPERIMENTS: The measurement set up was almost as same as that applied in the previous work [1]. Uranium (U)-Aluminum (Al) alloy samples was irradiated by the pulsed neutron and the γ ray was measured with a HPGe of the 35% relative efficiency. Data of pulse height of a signal in the HPGe and timing of the signal were accumulated event by event for OOPS. The length of the OOP time region for a neutron pulse is $(1/F - d)$ where d is the time required for decay out of the thermal neutrons. d should be longer than 16 ~ 20 ms [1] but longer d is not preferable since $(1/F - d)$ becomes less. The summation of the time length per second for OOPS counting is $F(1/F - d)$ and the neutron fluence per second on the U-Al sample is proportional to F. We chose F = 30 Hz since an index for γ ray count, $F^2(1/F - d)$, is larger for d = 16~20 ms, as shown in Fig. 1.

RESULTS: The measured spectrum in the OOP time region is shown in Fig. 2. Here, d = 20 ms. The measurement time was 17 hours. Thanks to the survey of F, counting statistics has been gradually enhanced compared to the previous work [1] so that the discrete peaks have been distinguished for γ ray energy region from 1.0 to 5.5 MeV. Based on the JENDL/FPY&FPD-2011 data [2], activities of the FPs were calculated by solving Bateman's formular and intense γ rays from the FPs were selected. By comparing the selected intense γ rays and the measured spectrum, the FPs radiating the γ rays are identified as shown in Fig. 2.

REFERENCES:

[1] Y. Nauchi *et al.,* KURNS Progress Report 2022 (2023) PR8-4.
[2] J. Katakura, JAEA-Data/Code 2011-025, 2012.

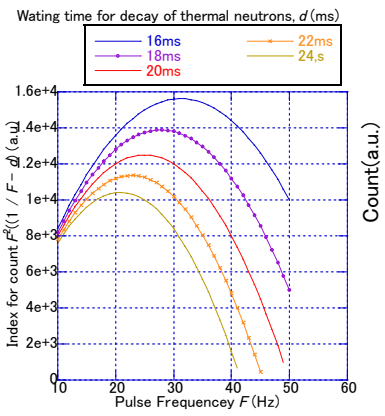

Fig. 1. Optimum Pulse frequency .

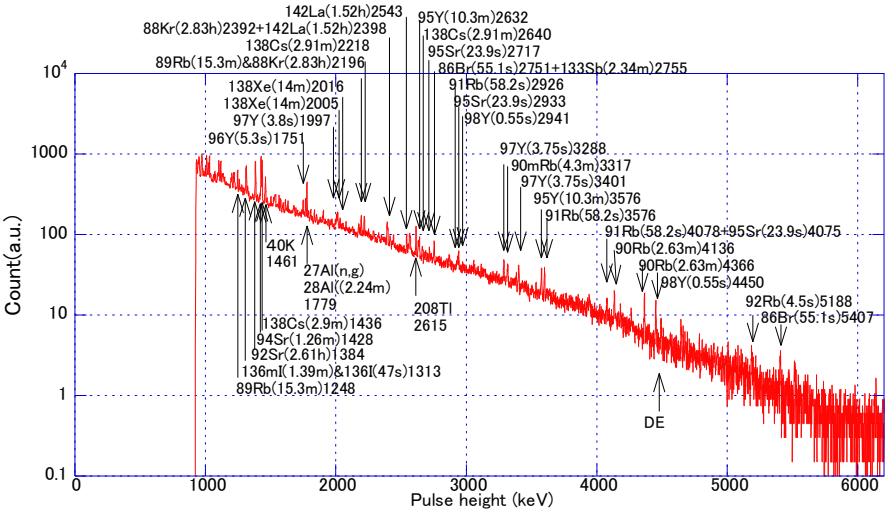

Fig. 2. Measured γ ray spectrum in OOP time region.

External Neutron Source for Non-Destructive Analysis of Fuel Debris (2)

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INTRODUCTION: As a non-destructive quantification technique of ^{235}U enrichment of irradiated nuclear material, neutron induced gamma ray spectroscopy (NIGS) has been studied [1]. For NIGS, we need an external neutron source. In the previous work, we had measured the γ ray background (BG) spectra inherent to three types of neutron sources, ^{252}Cf, ^{241}Am-Be, and a fusion neutron source in an inertial electrostatic confinement device (IEC source), by employing BGO scintillator. Among them, the IEC source is considered preferable since BG γ rays are not so intense in the energy region from 2.7~5.5 MeV where signature γ rays of ^{235}U, ^{238}U and ^{239}Pu are radiated [2]. In the current work, the BG γ rays were measured with good energy resolution.

EXPERIMENTS: Schematic view of the measurement is given in Fig. 1. Uranium (U) metal plates were loaded in rectangular aluminum pipe column with polyethylene plates. They were irradiated by neutrons from the ^{252}Cf or the IEC source. γ rays are measured with a HPGe of the 35% relative efficiency. From the U sample, γ rays of energy less than 100 keV was intense. To shield them, lead (Pb) plate and Pb-Bismuth (Pb-Bi) plates were placed. To increase the neutron flux in the U plates, polyethylene blocks and plates were placed surrounding the U plates. The measured pulse height was calibrated to energy referring ^{35}Cl(n, γ) γ ray spectrum measured additionally.

RESULTS: The measured pulse height spectra were shown in Fig. 2. With the ^{252}Cf source with intensity of 3 x 10^5 n/s, ^{238}U(n,γ) 4060 keV γ ray peak was identified within 2.5 hours of measurement. In the measurement, peaks corresponding to the neutron separation energy of $^{71, 74, 75}$Ge were measured. In the case where the U sample was irradiated by neutron from the IEC source, the neutron flux in the sample is less. Although we confirmed increment of count rate from 2.3 to 4 MeV mainly by the fission events, we could not identify the 4060 keV γ ray peak within 2 hours of measurement. When the IEC source was employed, ^{56}Fe(n,γ) 7631 and 7645 keV γ rays from the wall of the IEC device were identified. However, the count rates of them were less than those of γ rays from the $^{71, 75}$Ge(n,γ) reactions. That means major BG γ rays in this geometry were the neutron induced events in HPGe. To identify the ^{238}U(n,γ) 4060 keV γ ray peak with the IEC source, more than a hundred hours of measurement would be required in the current geometry.

REFERENCES:

[1] Y. Nauchi, H. Ohta *et al.,* Proc. ICNC2023, Sendai, Oct. 2023.
[2] Y. Nauchi *et al.,* KURNS progress report 2022, PP8-5.

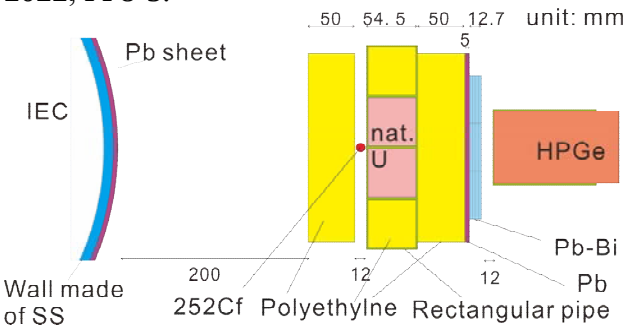

Fig. 1. Horizontal geometry.

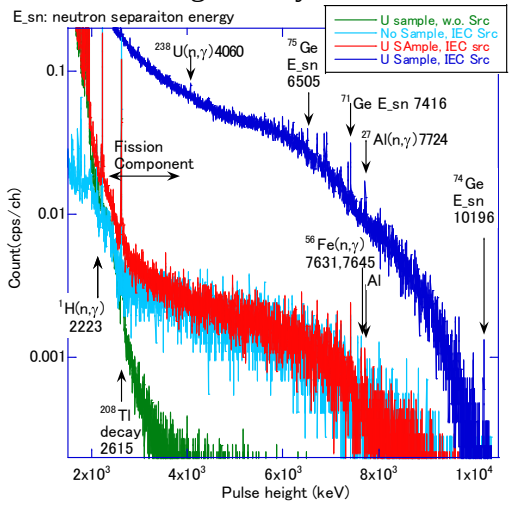

Fig. 2. Measured γ ray spectra.

Study of Non-destructive Analysis Method for Fuel Debris using Neutron Resonance Absorption

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INTRODUCTION: In the units 1 to 3 of Fukushima Daiichi Nuclear Power Plant (1F), the fuel, the metal cladding and the control rods were melted and large amount ware re-solidified in the bottom of the pressure vessel as fuel debris. In the decommission plan of 1F, the fuel debris will be collected and analyzed in the near future. Non-destructive nuclide assay is one of the important issues for nuclear material accountancy. We have studied the Neutron Resonance Densitometry (NRD) as a method of non-destructive nuclear assay. In the study, we proposed to apply a self-indication method to analysis of the fuel debris [1]. In the self indication method, an indicator consisting of the target nuclide is placed at the neutron beam downstream from a sample. The transmitted neutron though the sample can be measured indirectly by detecting the reaction products from the indicator with the neutron time-of-flight (TOF) method. We carried out the verification experiment.

EXPERIMENTS: The experiment was performed at the Institute for Integrated Radiation and Nuclear Science, Kyoto University (KURNS-LINAC). The linac was operated with a repetition rate of 200 Hz, a peak current of about 5 A, and an electron energy of about 30 MeV. We used a flight path in the direction of 135 degree with respect to the linac electron beam. A Cd sheet of 0.5 mm in thickness was inserted into the TOF beam line to suppress overlap of low energy neutrons due to the previous pulse. A natural uranium sheet as a indicator was set in the neutron beam line at a distance of about 12 m from the neutron target. Capture gamma-rays from the indicator were measured by a 4π $Bi_4Ge_3O_{12}$ spectrometer. On the other hands, three kinds of uranium samples with different thickness were set in the upper beam.

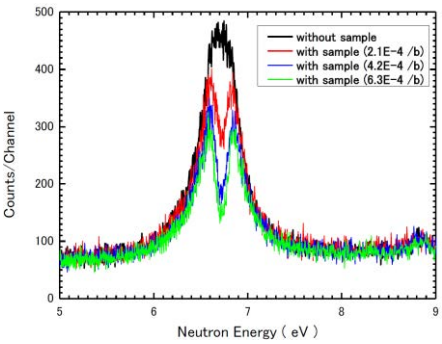

Fig. 1 TOF spectra for the 6.7-eV resonance of ^{238}U with and without the samples.

RESULTS: Figure 1 shows the TOF spectra for the 6.7–eV resonance of ^{238}U. The reduction of the resonance peak of the indicator was observed by inserting the sample in the upper beam. The reduction ratio has a relationship with the areal density of ^{238}U in the sample. We estimated the reduction ratio by using a Monte Carlo simulation code MCNP-6.3 with JENDL-5. The comparison between the experimental and the calculated reduction rates are shown in Fig. 2. The reduction ratios estimated by simple calculation reproduce the experimental ones. It was confirmed that the estimation of the reduction rate can be used for the determination of the areal density of fuel material.

In the future, we are going to perform the verification experiment using a realistic sample to simulate the fuel debris.

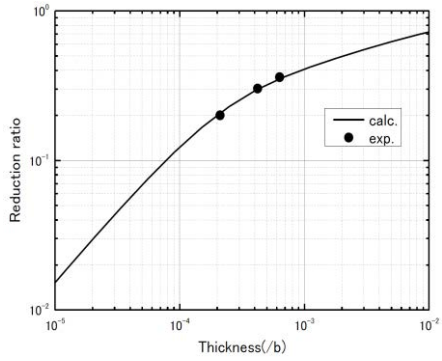

Fig. 2 Comparison between the experimental and the calculated reduction rates for the 6.7-eV resonance of ^{238}U.

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[1] J. Hori *et al.,* EPJ Web of Conferences **146** (2017) 09042 .