I-1. PROJECT RESEARCHES

Project 5

PR5

Fundamental Resratch 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.

R6P5-1: Neutron flux measurement for KUR facility activation modeling (2) (T. Sano *et al*., Kindai Univ.)
R6P5-2: Measurement of the amount of activation of structures around KUR (K. Takamiya *et al*., Kyoto Univ.)
R6P5-3: Fundamental Research about Radioactive Contamination Survey on Nuclear Reactor Facility - Elution tests with the irradiated KUR stack core - (S. Fukutani *et al*., Kyoto Univ.)
R6P5-4: Neutron Capture Cross-Section Measurement at TC-Pn in KUR for Holmium among Nuclides in Decommissioning (S. Nakamura *et al*., JAEA)
R6P5-5: Identification of Nuclear Material in Spent Fuel (3) (Y. Nauchi *et al*., Central Research Institute of Electric Power Industry)
R6P5-6: External Neutron Source for Non-destructive Analysis of Fuel Debris (Y. Nauchi *et al*., Central Research Institute of Electric Power Industry)
R6P5-7: Study on Non-destructive Analysis Method for Fuel Debris using Neutron Resonance Absorption (J. Hori *et al*., Kyoto Univ.)

MAIN RESULTS and CONTENTS of this REPORT:

T. Sano *et al*. (R6P5-1) evaluated the neutron flux distribution in the KUR sub-pile room during 5-MW operation for the decommissioning of KUR by an activation experiment and numerical calculations.
K. Takamiya *et al.* (R6P5-2) measured the activity concentration of ^{60}Co for the stainless steel screws used in the B-2 experimental hole irradiation apparatus. A clear correlation was observed between the neutron dose and the radioactivity of ^{60}Co.
S. Fukutani *et al*. (R6P5-3) conducted elution tests with the irradiated KUR stack core. The results of elution with 0.1 N nitric acid solution were obtained.
S. Nakamura *et al*. (R6P5-4) performed the activation experiments by using the thermal column pneumatic tube (TC-Pn) of KUR. The thermal-neutron capture cross-section for production of ^{166m}Ho with the half-life of 1133 years was obtained.
Y. Nauchi *et al*. (R6P5-5) measured FPs of half-lives from 0.5 s to a few hours by way of OOPS (out of phase event spectroscopy) technique at the pulsed neutron source in the KURNS-LINAC. We decreased the lower threshold energy of measurement from 950 keV in the previous work.
Y. Nauchi *et al*. (R6P5-6) studied enhancement of the count rate of the γ ray and reduction of background using a fusion neutron source in an inertial electrostatic confinement device (IEC source) as an external source.
J. Hori *et al*. (R6P5-7) performed the verification experiment with a self-indication method as a non-destructive assay at the pulsed neutron source in the KURNS-LINAC. It was confirmed that the method using a high-enriched uranium sheet as an indicator makes it possible to determine the areal density of ^{235}U contained in a natural uranium sample.

PR5-1

Neutron flux measurement for KUR facility activation modeling (2

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INTRODUCTION: For the decommissioning of the KUR, it is important to evaluate the amount of radio activation materials such as the experimental equipment, the biological shield and so on. In general, the evaluation of the activation dose is mainly based on numerical calculations. In numerical calculations, the neutron spectrum and absolute neutron flux are important input information. However, the absolute values often differ by several 10% or more between the calculated and measured values. With that background, the research project has been conducting neutron flux measurements on the surface of biological shields and constructing a numerical calculational model [1]. In the FY2024, the neutron flux at the sub-pile room in the KUR was measured by the radio-activation foils and detail calculation of neutron flux distribution of the KUR facilities.

EXPERIMENT: Neutron flux measurement in the sub-pile room were using by Au-197 foils. An Au-197 foil sample was 10 mm in diameter and 0.05mm thickness (approximately 75 mg). The foils were placed at 4 locations around the primary cooling water outlet piping in the sub-pile room shown in Fig.1. Neutron irradiation was conducted from January 21 to 23 in 2024, and gamma rays (411 keV) emitted from Au-198 were measured using a Ge detector.

CALCULATION: Neutron flux calculations in the KUR facilities 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.

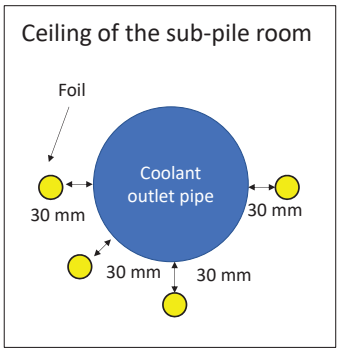

Fig.1 Location of Au foils setting

RESULTS: The averaged counting rate was 0.015 cps by the Ge detector. Since the reactor power during the irradiation period was 46 hours for 1 MWt and 6 hours for 5 MWt, the neutron flux evaluation was performed by dividing the counting rate by 1:4 as an approximation. In addition, the neutron flux can be considered as approximate thermal flux. As the results, the thermal neutron flux was $1.11 \times 10^4\ (n/cm^2/sec)$ at the 1 MWt and at the $5.56 \times 10^4\ (n/cm^2/sec)$ at the 5 MWt. In the numerical calculations, the thermal neutron flux in the sub-pile room was $3.0 \times 10^4\ (n/cm^2/sec)$ at the 5 MWt. The experimental and calculated values agreed by an order of magnitude.

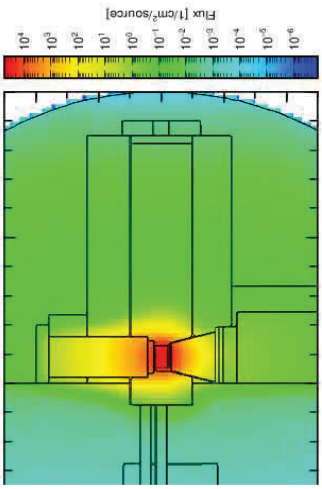

Fig.2 Calculated neutron flux distribution (relative value)

REFERENCES:

[1] T. Sano *et al.*, KURNS-Progress report 2022, (2023) pp.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.

PR5-2

Measurement of the amount of activation of structures around KUR

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INTRODUCTION: Unlike commercial nuclear reactors, safety measures for decommissioning research reactors require consideration of the activation of various experimental equipment such as experimental holes and irradiation holes. To provide basic data for safety assessments during the removal of these experimental structures, this study aimed to measure the amount of radioactive material present in some of these structures. To obtain a clear correlation between the actual measured values and the history of exposure to neutrons, the target structural materials were selected based on the condition that (1) there was no movement during the time they were installed, and (2) it was possible to estimate the irradiated neutron dose. The irradiation apparatus of the B-2 experimental hole met these criteria because its installation period is clear, and existing simulation-based calculated values and actual measured values obtained by activation analysis are available for the neutron dose within the experimental hole. Therefore, the irradiation apparatus of the B-2 hole was selected as the subject of this study.

EXPERIMENTS: The B-2 experimental hole irradiation apparatus was introduced as one of KUR's irradiation facilities in March 2012 and was removed in March 2021. This device was able to irradiate neutrons on large samples or liquid samples by transporting the truck with the samples from the adjacent laboratory into the experimental hole. A rail was installed within the inner sleeve of the experimental hole to transport the truck, and stainless steel screws were used to fix the rail. The position of these screws was precisely located using the apparatus drawings. Therefore, these screws were selected as the material for evaluating the amount of activation in this study. Six screws were collected at 30 cm intervals, starting from a range of 135 cm from the edge of the rail (i.e., at 135, 165, 195, 225, 255, and 285 cm from the edge). Gamma-ray spectra were subsequently measured using a Ge-semiconductor detector GC4020 (Canberra).

RESULTS AND DISCUSSION: As a result of a gamma-ray spectrum analysis, a photo-peak of ^{60}Co was observed, and ^{60}Co radioactivity was quantified by comparing it with the measurement results of a standard source of ^{60}Co. Figure 1 shows the correlation between the sample position and the radioactivity of ^{60}Co. The squares in the figure show the neutron doses previously estimated by the gold activation method. A clear correlation was observed between the neutron dose and the radioactivity of ^{60}Co. These results showed that by collecting a part of the structural material with a clear installation history among the experimental equipment and structures around the reactor, it was possible to infer the irradiation history of neutrons by measuring the radioactivity, and it was effective in assessing the effects of radiation exposure due to activation during decommissioning.

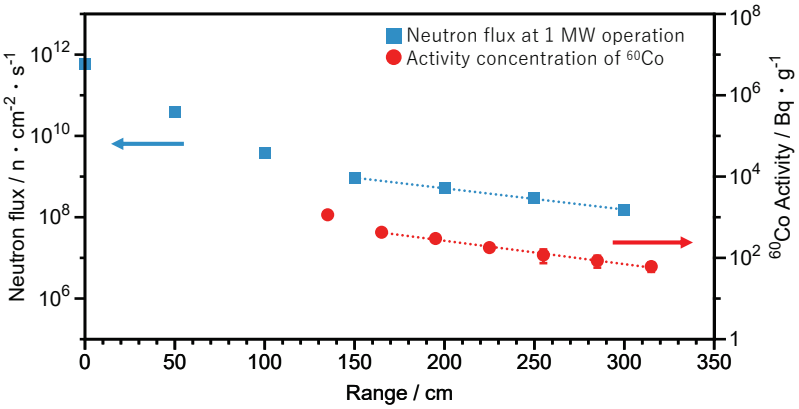

Figure 1 The correlation between the range from the edge of the rail and the activity concentration of ^{60}Co.

PR5-3

Fundamental Research about Radioactive Contamination Survey on Nuclear Reactor Facility - Elution tests with the irradiated KUR stack core -

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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. We reported stable nuclide concentration in the KUR stack core [1] measuring by INAA [2]. In this study, we conducted elution tests with the irradiated KUR stack core.

EXPERIMENTS: The KUR stack core samples, which was irradiated by neutron at KUR (Pn-2, 5MW, 1hour) were placed in a 15 mL centrifuge tubes, and ultrapure water or 0.1 N nitric acid solution was added. The solid-liquid ratio was 1:10, and the centrifuge tubes were stirred for 6 hours using a magnetic stirrer. At this time, a wire was used to make a two-layer structure to prevent contact between the stirrer and each core. After the agitation was completed, filtration was performed using a 0.45 μm disposable membrane filter. The filtrate was collected in a U-8 container. The obtained measurement samples were measured for nuclides using a Ge semiconductor detector (ORTEC GEM30-70) and a multichannel analyzer (SEIKO EG&G MCA7600). The measurement time was 80,000 seconds or longer. The radioactivity of the eluted nuclides was calculated from the measurement results.

RESULTS: Radionuclides were hardly detected in the filtrate when the elution tests were performed with ultrapure water. Therefore, only the results of elution with 0.1 N nitric acid solution were shown in Table 1.

Table 1 Elution rate of each nuclide from core 1 and core 2

nuclide	Core 1 [%]	Core 2 [%]
Ce-141	16 ± 0.6	23 ± 0.9
Ce-144	3.9 ± 0.3	3.7 ± 0.5
Co-60	20 ± 0.1	39 ± 0.3
Cr-51	9.7 ± 0.3	32 ± 1.0
Eu-152	15 ± 1.1	24 ± 2.0
Fe-59	5.9 ± 0.2	15 ± 0.3
Mn-54	15 ± 0.4	25 ± 0.7
Sb-124	6.1 ± 0.5	16 ± 1.3
Sc-46	5.5 ± 0.0	17 ± 0.1
Zn-65	25 ± 0.2	45 ± 0.4

Elution rate (%) = (Radioactivity of eluted nuclide [Bq]) / (Radioactivity of nuclide in the core sample [Bq]) × 100

REFERENCES:

[1] S. Fukutani *et al.,* KURNS Prog. Report 2022, (2023) 72.
[2] S. Fukutani *et al.,* KURNS Prog. Report 2023, (2024) 45.

PR5-4

Neutron Capture Cross-Section Measurement at TC-Pn in KUR for Holmium among Nuclides in Decommissioning

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INTRODUCTION: The present study is an attempt to measure accurate thermal-neutron capture cross-sections for nuclides of concern in decommissioning. From the viewpoint of clearance level [1] in decommissioning, we have performed the cross-section measurements by an activation method using the thermal column pneumatic tube (TC-Pn) of KUR [2-6]. The present work selected ^{165}Ho nuclide and measured its thermal-neutron capture cross-section for production of ^{166m}Ho with the half-life of 1133±8 years.

EXPERIMENTS: Two pieces of natural Ho foils were prepared with a purity of 99.9%, a diameter of 4mm, a thickness of 12.5 μm, and a weight of 15 mg. A gold-aluminum alloy wire, a cobalt foil and a Mo foil were used to monitor the neutron flux at the irradiation position. **Figure 1** draws a rough sketch of the target samples. The target was irradiated for 5 hours under 5-MW operation of KUR. After irradiation, the Ho samples and flux monitors were enclosed in a thin vinyl bag one by one. Gamma-rays emitted from each sample were measured with a high-purity Ge detector. A sample was placed at a distance of 110 mm from the front surface of the Ge detector. The γ-ray peak efficiencies of the Ge detector were measured in advance with a mixed γ-ray source and a gamma reference source of ^{152}Eu. Two Ho samples were measured together without overlapping each other. Gamma-rays emitted from ^{166g}Ho (26.8 hours) were measured with sufficient yields, therefore we also measured the cross section of the ^{165}Ho(n,γ)^{166g}Ho reaction. After ^{166g}Ho had decayed sufficiently, γ-rays from ^{166m}Ho were measured for 7 days. **Figure 2** shows a γ-ray spectrum of ^{166m}Ho. Four γ-rays originating from ^{166m}Ho were observed at 184, 280, 712, and 810-keV.

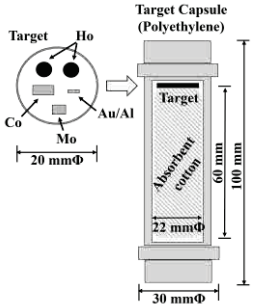

Fig.1 Target samples.

ANALYSIS AND RESULTS: The reaction rates of the Ho samples and flux monitors were calculated from their γ-ray yields. On the basis of Westcott's convention [7], the thermal-neutron flux was derived with the reaction rates of monitors. The cross sections for produced ^{166m,g}Ho were derived by dividing the reaction rates of Ho by the thermal-neutron flux. We tentatively obtained a value of 2.79±0.04 barn for the ^{165}Ho(n,γ)^{166m}Ho reaction, which was derived with an accuracy of 1.4% compared to the past reported value of 3.4±0.5 barn. As a by-product, we also obtained 61.2±0.6 barn for the ^{165}Ho(n,γ)^{166g}Ho reaction. Combining the cross sections for ^{166m}Ho and ^{166g}Ho presented 64.4±0.6 barn, which supports the past reported value of 64.4±2.8 barn by Time-Of-Flight method and the recently evaluated data of 64.69 barn and 64.4±1.2 barn within the limit of uncertainty.

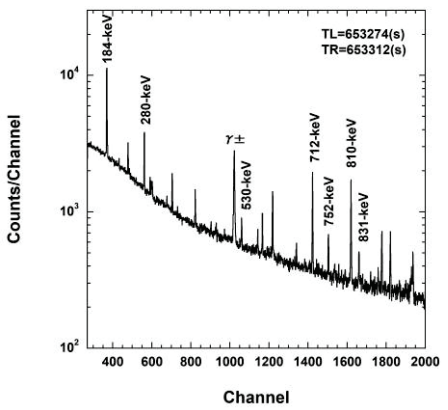

Fig.2 γ-ray spectrum of ^{166m}Ho .

REFERENCES:

[1] IAEA; 2004. (No. RS-G-1.7).
[2] S. Nakamura *et al.,* J.Nucl.Sci.Technol., **58**(10) (2021) 1061.
[3] S. Nakamura *et al.,* J.Nucl.Sci.Technol., **59**(11) (2022) 1388.
[4] S. Nakamura *et al.,* J.Nucl.Sci.Technol., **61**(11) (2024) 1415.
[5] S. Nakamura *et al.,* J.Nucl.Sci.Technol., **62**(3) (2025) 300.
[6] S. Nakamura *et al.,* J.Nucl.Sci.Technol., (2025) Published Online.
[7] C.H. Westcott *et al.,* P/202, Canada, **16** (1958) 70.

PR5-5

Identification of Nuclear Material in Spent Fuel (3)

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INTRODUCTION: γ ray emission data from short lived fission products (FPs) generated from ^{235}U had been measured to give reference data for delayed γ ray assay (DGA). We measured FPs of half-lives from 0.5 s to a few hours by way of OOPS (out of phase event spectroscopy) technique. The focused energy region was originally greater than 3 MeV for the purpose of DGA for spent nuclear fuel since γ ray spectroscopy for energy region less than 3MeV is impractical for such spent fuel. However, we had noticed γ ray intensity from FP is greater for lower energy region. In this work, we decreased the lower threshold energy of measurement from 950 keV in the previous work [1] to obtain full energy γ ray spectrum which is useful to validate decay heat data.

EXPERIMENTS: Measurement set up was almost same as that applied in the previous work [1]. The same Uranium (U) - Aluminum (Al) alloy sample was irradiated by neutrons periodically radiated from the tantalum target with frequency of 30 Hz.

The γ rays emitted from the sample were measured with the HPGe detector. The time spectrum of γ ray detection after pulsed electron was injected on the target is shown in Fig. 1. The prompt rays from the fission and the capture reactions appear in the time region from 0 to 16 ms. We shielded the HPGe detector with collimators of lead disks and boron doped polyethylene ones. We focused on the event in the time region from 20 to 33.3 ms. The region is out of phase (OOP) of the pulsed neutron so that γ rays were measured from radioactive decay of FPs and 235U. The ray spectra were measured on irradiation conditions for 18 hours.

Table 1. γ ray count rate for pulse height region.

Pulse height range (keV)	Count rate (cps)	error
70~210	99.97	0.25
210~950	187.62	0.07
950~3000	58.54	0.03
3000~	4.16	0.01

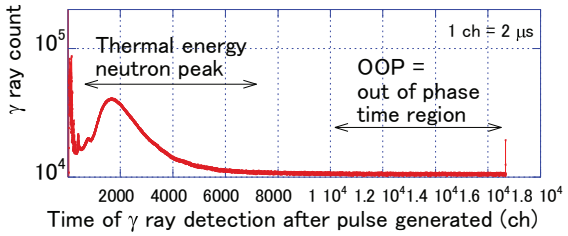

Fig. 1 Time spectrum of γ ray detection.

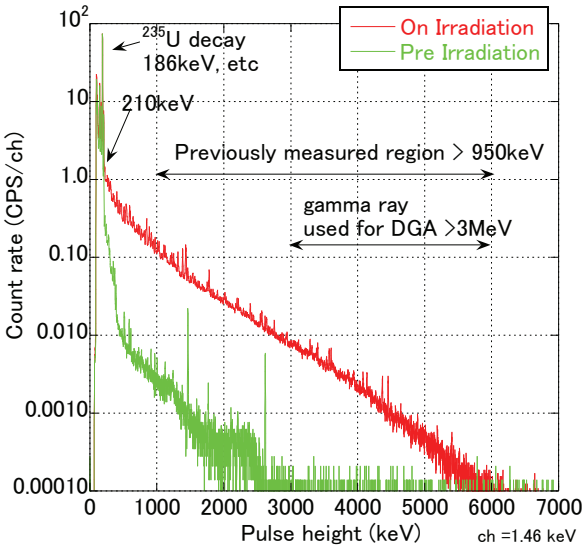

Fig. 2 γ ray spectrum in out of phase time region.

RESULTS: Measured spectra are shown in Fig. 2. By the sufficient collimation, we could reduce the ray detection threshold down to 70 keV. Below 210 keV, γ rays of energy 144, 163, 186, 205 keV, etc. from decay of ^{235}U are intense. The net count rate of the ray is shown in Table 1. The rate in the pulse height region extended in this work is 4.6 times larger than that in the previous work. Accord-ingly, these data are considered significant to validate total number of γ ray emission from FPs as well as the decay heat.

REFERENCES: [1] Y. Nauchi *et al.*, KURNS Progress Report 2023 (2024) 47.

PR5-6

External Neutron Source for Non-Destructive Analysis of Fuel Debris (3)

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INTRODUCTION: Neutron sources have been studied for neutron induced gamma ray spectroscopy (NIGS) for characterizing fuel debris retrieved from damaged cores [1]. A fusion neutron source from an inertial electrostatic confinement device (IEC source) is advanced since no significant γ ray emission is associated with the neutron emission. However, 4060 keV γ ray from the ^{238}U(n,γ) reaction had not yet been identified with a IEC source at KURNS [2]. In this work, enhancement of the count rate of the γ ray and reduction of backgrounds (BG) were studied.

EXPERIMENTS: NIGS with the IEC source was conducted in the measurement room of the KUCA building. Schematic view of geometry is shown in Fig. 1. The neutron source intensity was measured as 5×10^5 n /s. Metallic uranium (U) plates with the natural isotopic abundance were used. To enhance reaction rate of ^{238}U(n,γ), we made a unit cell which consists of a sheet of the U plate and two sheets of the polyethylene plates with 3.085 mm in thickness. The 5 units were loaded in a square shaped sheath made of aluminum. Polyethylene reflectors were placed on both sides of the units. The thickness of the reflectors was chosen to enhance the count rate of the 4060 keV γ rays. Sixteen sets of the U samples, polyethylene plates, and the sheath were employed, where the number of U plates was doubled compared to that employed in the previous work [1]. ^{238}U(n,γ) 4060 keV γ rays were measured with a HPGe detector. Dead time of it due to γ rays from ^{234m}Pa in the U plates is issue. To mitigate the dead time, 6 sheets of lead-bismuth plates were loaded at the end of the sheath. In the previous work, γ ray emission in the HPGe detector from neutron capture reactions of germanium isotopes was major BG [2]. To reduce BG, we placed lithium fluoride plates which absorb thermal neutrons without radiating γ rays. To avoid spectrum distortion due to high counting rate, shaping time of pulse signals by the linear amplifier was set to 2 μs.

RESULTS: Pulse height spectrum obtained by 3.5 hours of measurement is shown in Fig. 2. The pulse height was calibrated referring to 1001 keV γ ray from ^{234m}Pa and 2614.5 keV γ ray from ^{208}Tl assuming linearity of pulse height channel to energy. As the results, we identified 4060 keV γ ray peak above continuum fission prompt γ ray spectrum although counting statistics is not sufficient. However, the statistics can be enhanced just by extension of the measurement time up to a day. Accordingly, IEC neutron source is promising for NIGS for the fuel debris.

REFERENCES: [1] Y. Nauchi *et al.*, KURNS Progress Report 2022 (2023) 74.
[2] Y. Nauchi *et al.*, KURNS Progress Report 2023 (2024) 48.

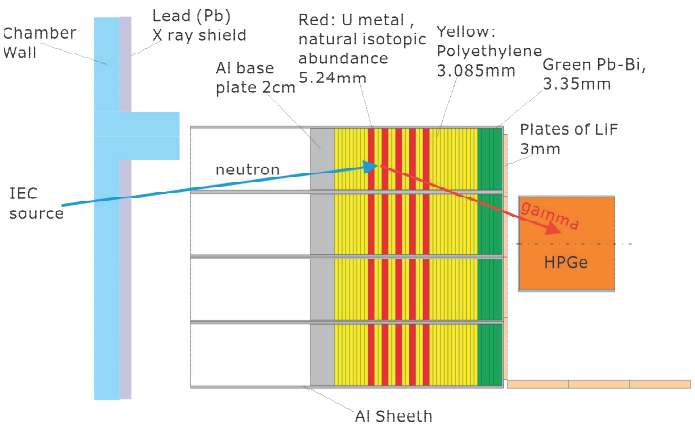

Fig. 1 Schematic view of measurement geometry.

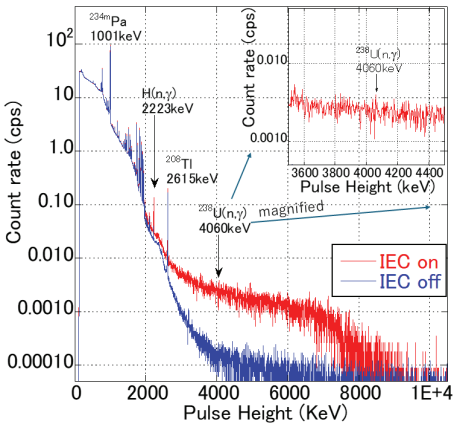

Fig. 2 γ ray spectrum from U irradiated by IEC n-source.

PR5-7

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 high-enriched uranium (HU) sheet was used as an indicator. The HU indicator was made of U_3O_8-Al alloy and the areal densities of ^{235}U and ^{238}U were 1.3×10^{-4} and 9.5×10^{-6} /b, respectively. Prompt gamma-rays from the indicator which set at the position of 12m neutron flight path were measured by a 4π BGO spectrometer. Another measurement with a natural uranium in the upper beam side was also performed. The sample was 1 cm long, 2 cm wide and 5.8 g in weight.

RESULTS: Figure 1 shows the comparison of TOF spectra for prompt gamma-rays from the HU indicator with and without the natural uranium sample. The resonances due to fission reactions of ^{235}U and capture reactions of ^{235}U or ^{238}U were observed. It is noted that the effective reaction rate of the 20.9-eV resonance of ^{238}U becomes to be comparable to that of the 19.3-eV resonance of ^{235}U in the case of HU indicator. By inserting the natural uranium sample, the decrease in peak area depending on the areal density for each nuclide was observed. We succeeded in assaying the areal density of ^{235}U in the natural uranium sample by the self-indication method.

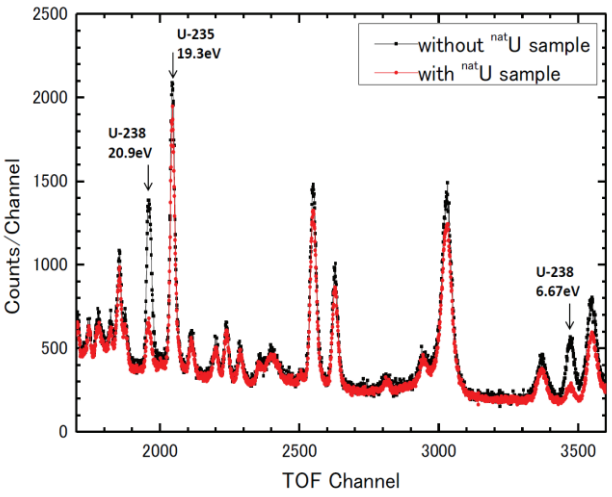

Fig. 1 Comparison of the TOF spectra for prompt gamma-rays from the HU indicator with and without natural uranium sample

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