CO2-1 Study on Neutron Beam Pulse width Dependence in the Nuclear Fuel Measurement by the Neutron Resonance Transmission Analysis

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INTRODUCTION: As a non-destructive analytical technique for nuclear material in the field of nuclear security and nuclear nonproliferation, a neutron resonance transmission analysis (NRTA) attracts attention of researchers [1]. A large electron beam accelerator is generally used as a neutron source of a NRTA system so far. It is important to downsize a NRTA system when it is deployed at various facilities such as a reprocessing one [2]. For this aim, we have developed a compact NRTA system which utilizes a D-T neutron generator [3]. Its pulse width of 10μs is much longer than that of a large electron beam accelerator. It is necessary to understand the influence of pulse widths on the NRTA measurement. Therefore, we conducted the experiments of the simulated nuclear fuel pin samples to evaluate how the NRTA measurement is influenced by the pulse width of neutron beam.

EXPERIMENTS: Experiments were performed in KURNS-LINAC (Kyoto University, Institute for Integrated Radiation and Nuclear Science - Linear Accelerator). A sample changer was installed at the neutron irradiation line. A Li-6 detector was located at the position of approximately 7 m from the neutron source, and was utilized to measure neutrons which transmit through a sample. The simulated fuel pellet sample was made from metallic powders of Ag (around 1%) and Al (around 99%). Numbers in parentheses indicate weight ratio of each powder. The pellet size is about φ 6.5mm x t 10mm. The six pellets were put in a stainless case to simulate a fuel pin. The energy of the irradiation neutron is determined by a Time of Flight (TOF) technique. In addition to the sample, metallic plates of W and In were inserted in a neutron beam to investigate pulse width effects as well as to determine the neutron energy. The thicknesses of the W and In plates are 0.5mm and 0.2mm, respectively. In this work, we used three pulse widths of the neutron beam of 0.1, 1 and 4 μs.

RESULTS: An example of a TOF spectrum measured is shown in Fig. 1. A resonance dip of ^{109}Ag at 5.19 eV is observed in the spectrum. The Ag resonance dip is able to measure in the other pulse widths. The resonance energies of ^{235}U, ^{238}U and ^{239}Pu are around 5eV. Therefore, it is confirmed that NRTA can be utilized for the measurements of nuclear fuel materials in the fuel pin sample.

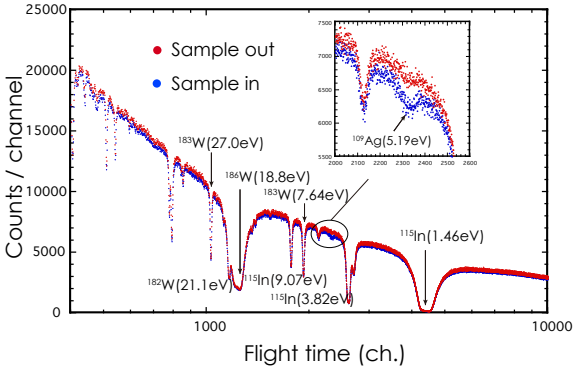

Fig. 1. TOF spectrum with pulse width 0.1μs

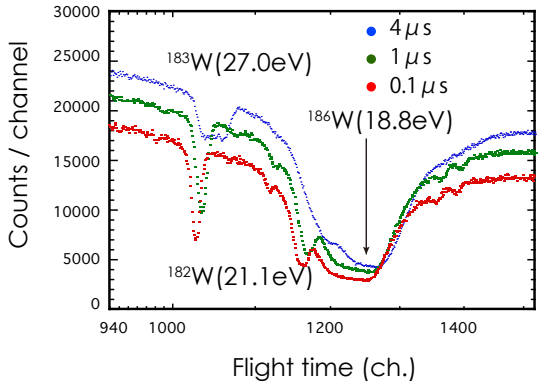

Fig. 2. The TOF spectra of individual pulse widths

Figure 2 shows the variation of TOF spectrum in the pulse width change. As can be seen, the dip of the TOF spectrum shifts towards low energy, with pulse width changed to a longer one. Resonance dips of ^{182}W and ^{186}W are not well separated in the 4 μs spectrum. Furthermore, the ^{183}W resonance dip is strongly skewed in the 4 μs spectrum. These mean that the resolution of the energy is worse with longer pulse width, and it would be difficult to apply a resonance analysis to the dips in the higher-energy region over approximately 20 eV with a longer pulse width of 4 μs. In this work, we confirmed that neutron pulse width affected the NRTA measurement of the fuel pin sample. On the basis of this work, we will be able to quantify the effects of long-pulse width in a resonance analysis.

ACKOWLEDGEMENTS: This research was implemented under the subsidiary for nuclear security promotion of MEXT.

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CO2-2 Measurement of Doppler Effect by Small Accelerator Neutron Source (II)

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INTRODUCTION: In order to reduce TRU, the research and technology development entitled as "TRU burning fast reactor cycle using uranium-free TRU metal fuel" have been started Japan at October 2014 [1]. The feature of the fast reactor is high content TRU and Zr without uranium in the fuel alloy so that additional TRU is not produced. On the other hand, uranium-free TRU metallic fuel leads to the reduction of the Doppler reactivity. Thus, the utilization of fuel alloy such as Mo and Nb instead of Zr is considered as one of the countermeasures [2]. As the Doppler effects depend on the magnitude of self-shielding at the resonances, it is important to verify the Doppler effects at each resonance of the fuel alloy (Mo or Nb) materials to evaluate the feasibility of the uranium-free TRU metallic fuel. Therefore, we have performed the measurement of the Doppler effects for Mo sample by Time-of-flight (TOF) method with the KURRI-LINAC pulsed neutron source since 2016 [4, 5].

EXPERIMENTS: We measured the Doppler effects of a Nb metal sample using a pulsed neutron source at the 46 MeV electron linac in the Integrated for Radiation and Nuclear Science, Kyoto University. In the experiment, neutron capture rates in the Nb sample at 300 K and 600 K were obtained by prompt gamma-ray measurement with the TOF method. The Nb sample was placed in the center of a heating device at a distance of 10 m from the Ta target. The surface temperature of the sample was observed by thermoelectric couple and controlled to be constant during the irradiation by a glass-heater. Two kinds of samples with different thickness of 1.0 mm^t and 3.0 mm^t were prepared to identify the neutron self-shielding effects for each resonance. Those samples had cross sections of 2.0×2.0 cm^2. The measurements with thick and thin samples, no sample (blank run) at 300 K were carried out. The experimental conditions are shown in table 1.

Table 1 Experimental conditions

Sample	Temperature (K)	Measurement time (h)
3.0 mm^t	300	28.0
3.0 mm^t	600	30.8
1.0 mm^t	300	11.7
1.0 mm^t	600	9.9
Blank	300	7.2

RESULTS: The measured TOF spectra with 1.0 mm^t and 3.0 mm^t Nb samples at 300K are shown in Fig. 1. The main resonances of Nb-93 were clearly observed in the energy range from 10 eV to 10 keV. For further analysis, counting statistics observed in TOF spectrum were improved by the JSF70 group structure [6] corresponding to the lethargy width of 0.25. Table 2 shows the absorption rate ratio of 3.0 mm^t sample by Doppler effect. The absorption rate ratio defined as the absorption rate at 600 K divided by it at 300 K was obtained for each energy group. As the results, the absorption rate ratio were 1.16 ± 1.9 % at the 41th group, 1.09 ± 2.3 % at the 42th group, 1.15 ± 1.3 % at the 44th group, 1.04 ± 1.6% at the 46th group and 1.06 ± 4.9 % at the 51th group.

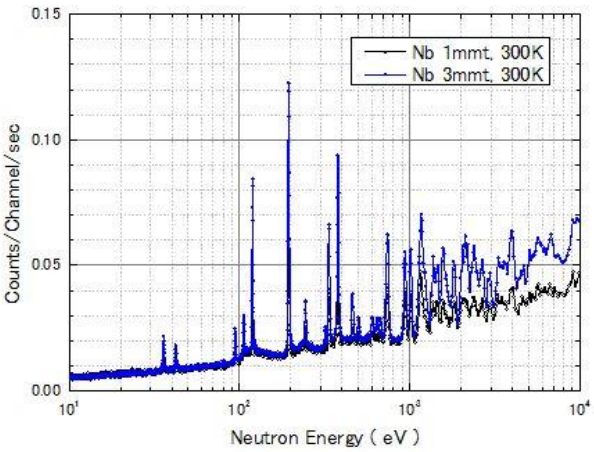

Fig1. Measured TOF spectrum

Table 2 Absorption rate ratio of 3.0 mm^t sample by Doppler effect

Energy group	Resonance (eV)	Absorption rate ratio
41	378.5	1.16±1.9%
42	335.4	1.09±2.3%
44	193.8	1.15±1.3%
46	105.9, 119.1	1.04±1.6%
51	35.9	1.06±4.9%

This study is being performed under the contract with the Ministry of Education, Culture, Sports, Science and Technology of Japan (MEXT) in the framework of MEXT's Nuclear System Research and Development Program.

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CO2-3 Study on the Non-destructive Nuclide Assay for Nuclear Materials with a Self-indication Method

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INTRODUCTION: A fast reactor system with trans-uranium (TRU) fuel containing minor actinides (MA) is considered as one of options for reduction the high-level radioactive waste from the nuclear power plants. It is important to test their integrity to confirm the fuel safety. Non-destructive nuclide assay is a key technology for nuclear material accountancy. Neutron Resonance Densitometry (NRD) is a non-destructive nuclide assay technique applicable to quantity nuclear materials in the fuel. For the TRU fuel having high radioactivity, a self-indication method [1] is adapted for the identification and quantification of nuclides in the fuels. 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 thorough the sample can be measured indirectly by detecting the reaction products from the indicator with the neutron time-of-flight (TOF) method. The self-indicator is a transmission neuron detector that has high efficiency around the objective neutron resonance energies of the target nuclide, enabling us to quantify effectively the amount of resonance absorption of the target nuclide. Moreover, it is not easily affected by the decayed gamma-rays from the fuel. We carried out the quantitative examination of self-indication method for a nuclear material with highly enriched and depleted uranium-aluminum alloys.

EXPERIMENTS: The experiment was performed at the 46-MeV electron linear accelerator in Institute for Integral Radiation and Nuclear Science, Kyoto University. The linac was operated with a repetition rate of 300 Hz, a pulse width of 100 ns, a peak current of about 5 A, and an electron energy of about 35 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 also inserted into the TOF beam line to suppress overlap of low energy neutrons from the previous pulse due to the high frequency of the linac operation. A 4π $Bi_4Ge_3O_{12}$ (BGO) spectrometer was installed at the 12m flight path in the TOF beam line for the measurement of capture gamma rays from the indicator. A natural uranium sheet with the areal density of 7.36×10^{-3} (/b) was used as a indicator. Five samples with different thickness were prepared by combining high enriched and depleted uranium-aluminum alloys. TOF measurements with and without samples were performed, respectively.

RESULTS: The TOF spectra with and without samples are shown in Fig. 1. The resonances of ^{238}U were clearly observed at 6.67, 20.9, 36.7, 66.0, 80.7 and 103 eV. It can be seen that the reduction ratio of the resonance peak areas become larger as the areal density of ^{238}U increases. On the other hands, the relation curve between reduction ratio and the areal density was obtained by using Monte Carlo simulation for each resonance as shown in Fig. 2. As the results, the areal density of ^{238}U was obtained successfully by comparing the experimental reduction ratio with the estimated relation curves.

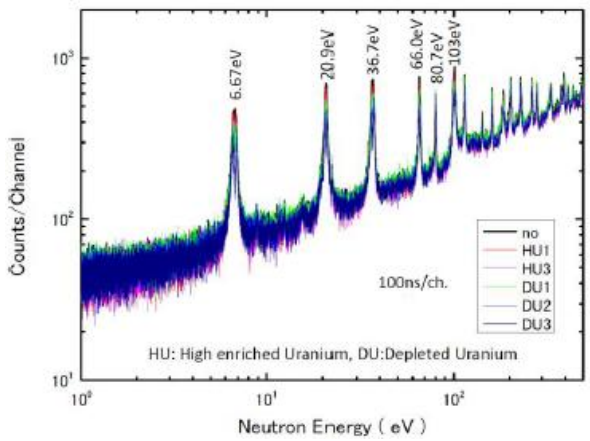

Fig.1 Comparison of TOF spectra with and without samples

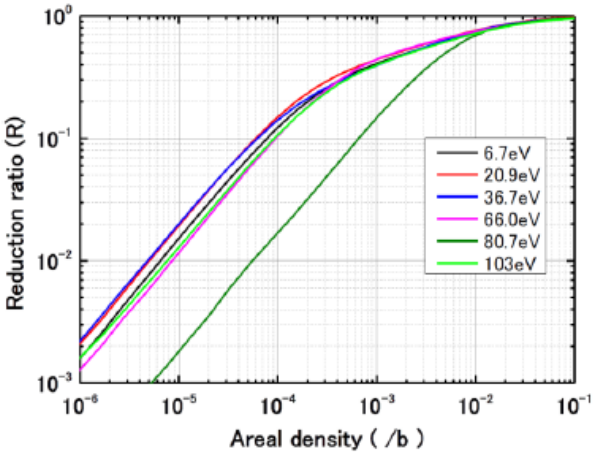

Fig.2 Relation between the reduction ratio and the areal density of the sample for each resonance of ^{238}U

Present study includes the result of "Development of Non-Destructive Methods Adopted for Integrity Test of Next Generation Nuclear Fuels" entrusted to the Kyoto University by the Ministry of Education, Culture, Sports, Science and Technology of Japan (MEXT).

REFERENCE:

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CO2-4

Development of Gamma Imager and Its Application to Identification of Nuclear Materials

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INTRODUCTION: Identification of a hidden or shielded gamma radioactive sources and nuclear materials is required in anti-terrorism measures. We are developing passive and active gamma spectroscopy combined with gamma-ray imaging for these applications. Compton gamma imager can measure energy of incident gamma-ray and its direction to the detector. We applied a portable and 4π direction sensitive Compton imager [1-2] to identify ^{10}B sample by prompt gamma-ray with neutron irradiation at the pulsed neutron source at Kyoto University Institute for Integrated Radiation and Nuclear Science Linear Accelerator (KURNS-Linac).

EXPERIMENTS: Figure 1 shows an experimental setup at the pulsed neutron source. A 3D pixel array CdTe detector with 1440 ch. was used as a prototype of the 4π sensitive gamma-ray imager. The prototype CdTe detector was located at a distance of around 12 m from the neutron source. In our previous work based on Monte Carlo simulation using EGS5, the prototype Cd detector would be capable to identify a source position even if the gamma-ray energy emitted from the source is lower than 400 keV. A ^{10}B sample in an aluminum disk was irradiated by the pulsed neutron on the beam line.

RESULTS: Figure 2 shows a typical measured energy spectrum from the irradiated ^{10}B sample. Peaks corresponding to prompt gamma-ray of ^{10}B (478 keV) and ^{113}Cd (558, 651 keV etc.) appear clearly in the spectrum. Typical reconstructed image of ^{10}B prompt gamma-ray on a projection sphere around the prototype CdTe detector is shown in Fig.3. Note that ^{10}B was used in a shielding material surrounding the outlet of the pulsed neutron beam (θ=50°, φ=0) in this setup. Whereas the position of the ^{10}B sample (θ=–90°, φ=0) was localized by the prompt gamma-ray imaging, background events on the image were rather high due to interference of prompt gamma-rays from ^{113}Cd in the CdTe detector. To suppress the background in prompt gamma-ray analysis using the CdTe detector, we investigated shielding effect of a lithium fluoride tile to ^{10}B prompt gamma-ray used as a shielding material for thermal neutrons without any productions of gamma rays by neutron absorption. One lithium fluoride tile was put in the front surface (θ = –90° plane) of the detector. Even after passing through the lithium fluoride tile, the resolution of ^{10}B prompt gamma-ray image had same as that without the tile.

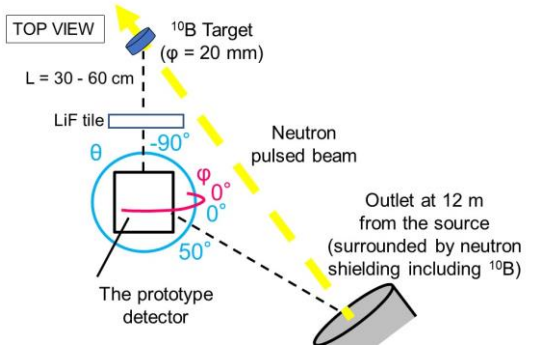

Fig. 1. Experimental setup at the pulsed neutron source.

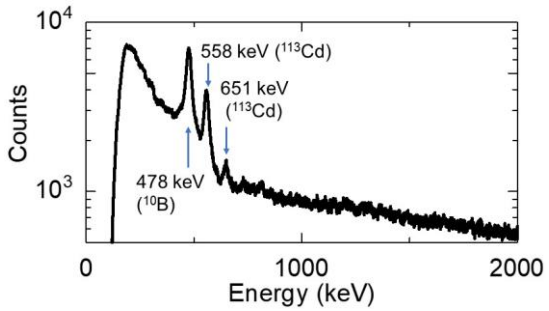

Fig. 2. Typical energy spectrum from the ^{10}B sample.

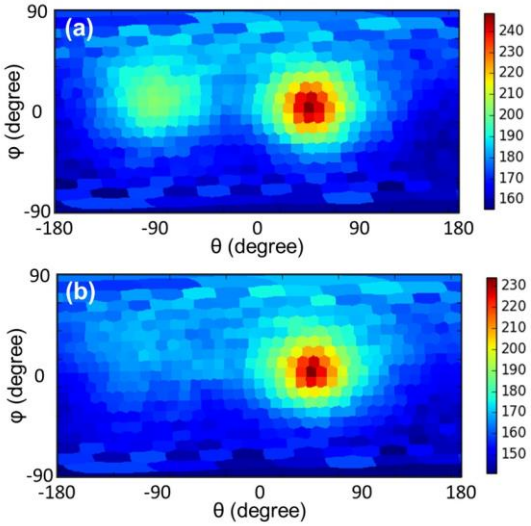

Fig. 3. Reconstructed images on the projection sphere (θ, φ), (a) with the ^{10}B sample and (b) with a dummy sample not including ^{10}B.

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CO2-5 Measurements of Epithermal Neutron Detector with a Boron Loaded Plastic Scintillator

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INTRODUCTION: Evaluation of neutron fluence and neutron dose equivalent for the epi-thermal neutron region is very important in work places with neutron sources or nuclear fuels as well as irradiation fields in a boron neutron capture therapy. In the present study, we have developed a precise epi-thermal measurement method that is not affected by nuclear reaction cross sections such as the $^{10}B(n,\alpha\gamma)$, $^{3}He(n,p)$ and $^{6}Li(n,\alpha)$ reac- tions. We experimentally evaluate the neutron detection system for absolute measurements of epi-thermal neutrons. The epi-thermal neutron detection system for absolute measurement is composed of a 50.8 mm diameter and 25.4 mm thick boron loaded plastic scintillation detector and two 50.8 mm diameter and 50.8 mm thick BGO scintillation detectors. In our previous experiments, a $^{6}Li_{6}{}^{nat}Gd^{10}B_3O_9$:Ce+ (LGB) scintilla- tor was used as a boron containing scintillator. However, the LGB scintillator is not yet commonly used. In the present study, we tried to demonstrate epi-thermal neutron measurements using the neutron detection system with a boron loaded plastic scintillator that is commonly used.

EXPERIMENTS: A collimated neutron beam was obtained by the photo neutron reaction using a water-cooled tantalum target at the KURRI Linac [1]. Fig1 shows the typical experimental arrangement. The boron loaded plastic scintillation detector was set at the center of beam line. The BGO scintillation detectors were placed on both side of the boron loaded plastic scintillation detector. For low energy neutrons, monoenergetic gamma rays with energy of 478 keV and alpha particles are produced in the plastic scintillation detector by the $^{10}B(n,\alpha\gamma)$ reaction. The alpha particles and a part of gamma rays are observed with the boron loaded plastic scintillator. The BGO scintillation detectors detects the rest of gamma rays. Characteristics of the neutron detection system were experimentally evaluated by means of the time-of-flight (TOF) method.

RESULTS: Fig.2 shows the two-dimensional plots of pulse heights of the boron loaded plastic scintillation detector and the BGO scintillation detector. In Fig.2, the area A indicates that mono energetic gamma rays with 478 keV are detected by the BGO scintillation detector. In this case, the gamma rays produced by the $^{10}B(n, \alpha\gamma)$ reaction do not occur any reaction in the boron loaded plastic scintillator. On the other hand, the area B indicates that gamma rays are detected by the BGO scintillation detector after the Compton scattering reaction of the 478-keV gamma rays in the boron loaded plastic scintilla- tor. We are developing a data analysis algorithm. The experimental results will be compared with experi- mental results of the previous studies.

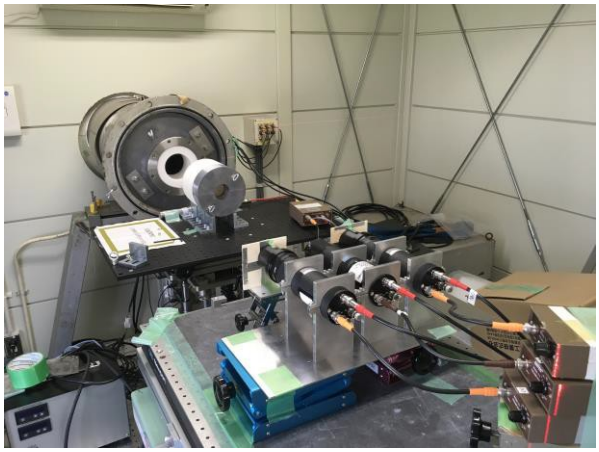

Fig. 1. Typical experimental arrangement in the KURRI linac facility.

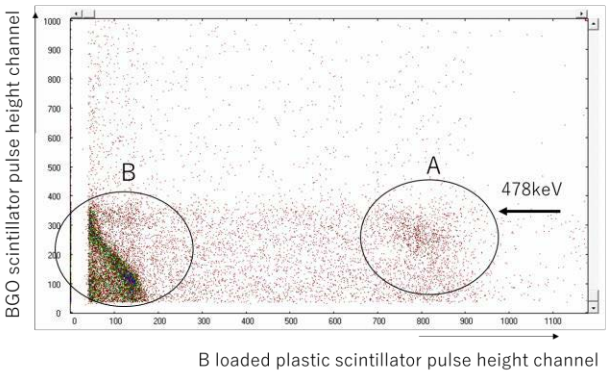

Fig. 2. Two-dimensional plots of pulse heights of the boron loaded plastic scintillation detec- tor and the BGO scintillation detector.

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CO2-6 Study of Isotope Separation via Chemical Exchange Reaction

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INTRODUCTION: By utilizing chemical separation method for calcium isotope effect in liquid-liquid extraction (LLE), an appropriate crown-ether (CE) can be used not only for separation of metal ions, but also for separation of isotope[1].

EXPERIMENTS: Isotopic enrichment occurs according to the following chemical exchange reaction:

$$^{40}Ca^{2+}_{(aq)} + {}^{48}CaL^{2+}_{(org)} \rightarrow {}^{48}Ca^{2+}_{(aq)} + {}^{40}CaL^{2+}_{(org)} \quad (1)$$

where L represents macrocyclic polyether(18-crown-6). A 20 ml aqueous solution (3M $CaCl_2$) and 200 ml organic solution (0.07M DC18C6 in chloroform) were stirred by a magnetic stirrer for 60 m at room temperature and separated. This LLE was iterated six times (1－6)[2]. The large isotope effect with mass dependence was obtained by the back-extraction (BE) method [2]. However the Ca concentration was heavily degraded to the order of 10^{-6} after the six iteration/multistage LLE and this low mass content cannot be realized for the actual enrichment of Ca with a ton scale refinement. Thus we developed a new LLE procedure with maintaining the Ca concentration (partition factor) as much as possible and also keeping the separation factor as large as possible at the same time, which is shown in Fig. 1. It is well known that the relation between a separation factor and a partition factor is in a trade-off relationship.It is also noted that we can easily reuse the CE solution having Ca inside its cavity by detaching Ca via pure water only. By comparison, in the case of CE resin we are required to use high content (～9M) of HCl [3]. It is quite important that we can reuse the valuable CE easily and many times in terms of a cost-effective enrichment.

RESULTS: The Ca concentration in aqueous and organic phase for the new LLE procedure was measured by AAS. We can obtain about 20 % partition factor and its Ca transfer between two phases can be confirmed in Fig. 2, not to the other medium such as chloroform.

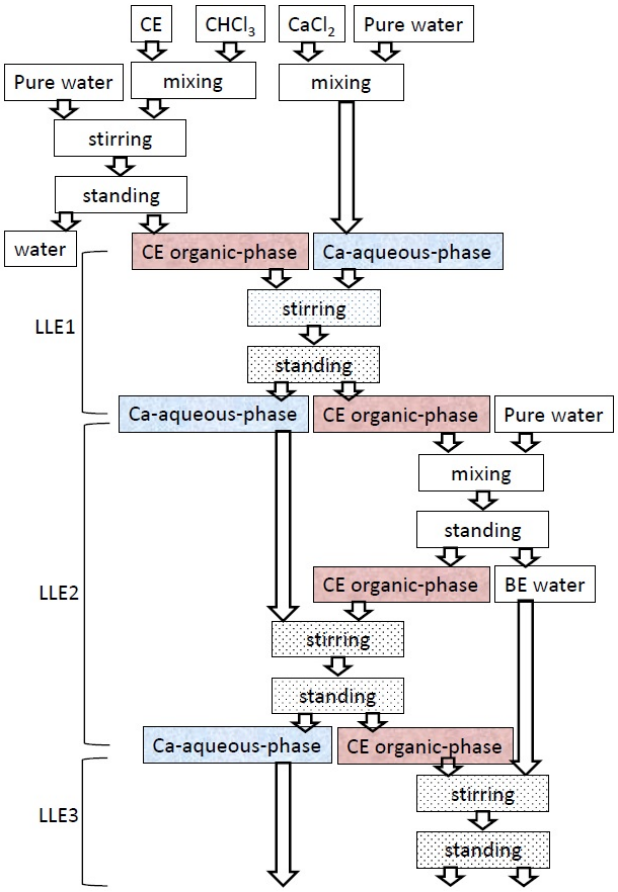

Fig 1. New LLE procedure with maintaining Ca concentration as much as possible and a separation factor at the same time, which allows to reuse CE via water only.

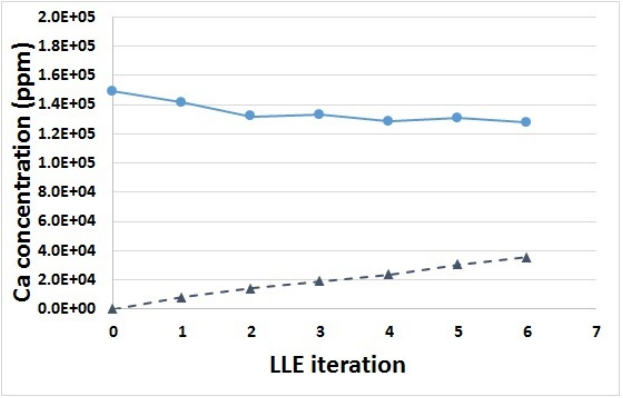

Fig 2. Ca concentration in aqueous phase (circle) and organic phase (triangle) for the process of LLE. Iteration is six times, zero is feed solution for $CaCl_2$ aqueous and crown-ether organic ($CHCl_3$) phase. : Preliminary.

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CO2-7 Reactivity Measurement of Accident Tolerant Control Rod Materials

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INTRODUCTION: As an effort to improve the safety of light water reactors (LWRs), various concepts of "enhanced accident tolerant fuels and core components" are being developed in many countries. The Central Research Institute of Electric Power Industry (CRIEPI) has been developing an accident tolerant control rod (ATCR) to which the novel neutron absorbing materials including rare-earth sesquioxides (RE_2O_3) are applied [1]. The ATCR improves the basic performance as a control rod such as the reactor shutdown margin and neutronic lifetime, and reduces the risk of re-criticality accident in any reactor conditions including severe accident (SA). For the ATCR candidate materials RE_2O_3-MO_2 (RE = Sm or Eu, M = Zr or Hf), a high temperature compatibility with iron or steel, which is a main component of current control rod cladding, and physicochemical stability under high temperature steam atmosphere were experimentally confirmed so far [2]. Furthermore, a reactivity analysis in the representative LWRs revealed that these candidate materials have enough reactivity worth comparable to or higher than the conventional neutron absorbing materials B_4C or Ag-In-Cd alloy [1]. In this study, the reactivity worths of RE_2O_3-MO_2 are measured to verify the analysis results in view of the importance of control rod functions such as power adjustment, shutdown and scram of reactor.

EXPERIMENTS: A critical core with thermal neutron spectrum (E3 core) was assembled in A-core of Kyoto University Critical Assembly (KUCA) as shown in Fig. 1. The unit cell of fuel assemblies is composed of a 93% enriched ^{235}U-Al alloy fuel plate of 2"×2"×1/16" and 3 polyethylene plates of 2"×2"×1/8"/plate. An Al-holder with an inner diameter of 1.2cm and a depth of 1.969cm that contains a mixed powder sample of RE_2O_3-MO_2 was loaded in the center of core and the reactivity worth of each sample was measured with the period method. Reactivity worth of each sample in E3 core was calculated using the continuous-energy Monte Carlo code MVP-3 with 2.58 billion neutron histories. The statistical error of reactivity calculation was less than 3pcm. The neutron cross section libraries were generated using JENDL- 4.0.

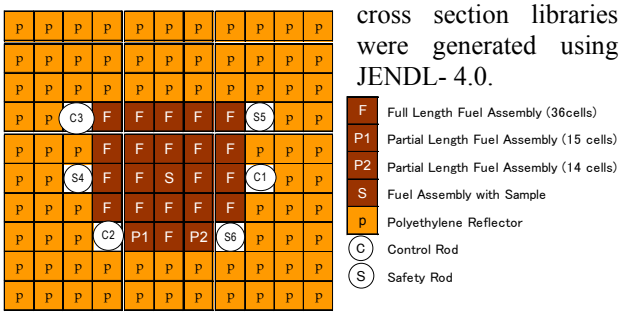

Fig. 1 Configuration of E3 core assembled in KUCA A-core.

RESULTS: The measurement results of reactivity for mixed powders of Sm_2O_3-MO_2 and Eu_2O_3-MO_2 are shown in Figs. 2 and 3, respectively. The self-shielding effect due to the strong neutron absorption of RE_2O_3 suppresses the change in reactivity worth even if excessive RE_2O_3 of 1.5-2.0g or more is loaded. Tables I and II summarize the differences between calculations (C) and measurements (E) of reactivity for Sm_2O_3-MO_2 and Eu_2O_3-MO_2, respectively. For all samples, the reactivity worth was predicted with high accuracy of less than 10pcm differences. From these results, validity of nuclear data of Sm, Eu, Zr and Hf in the thermal energy region was verified. As confirmed by the reactivity worth analysis in the representative LWRs [1], RE_2O_3-MO_2 is considered to be applicable as the alternative control materials.

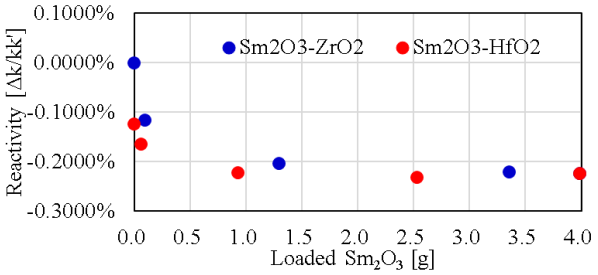

Fig. 2 Measurement results of reactivity for Sm_2O_3-ZrO_2 or Sm_2O_3-HfO_2.

Table I Differences between Calculations and Measurements (C-E) of Reactivity for Sm_2O_3-ZrO_2 or Sm_2O_3-HfO_2

Sm_2O_3[g]	0	0.058	0.93	2.5	0	0.093	1.3	3.4	4.0
HfO_2 [g]	6.7	7.0	5.6	3.1	0	0	0	0	0
ZrO_2 [g]	0	0	0	0	6.6	6.6	4.6	1.1	0
C-E [pcm]	6.4	8.6	5.8	2.9	3.4	3.2	-3.9	-2.3	3.4

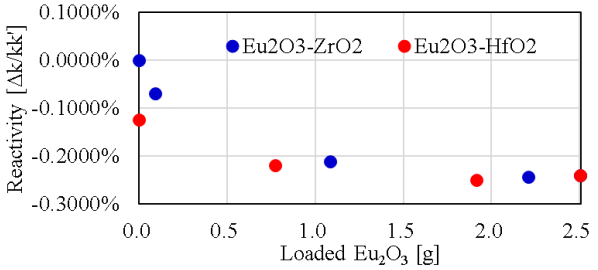

Fig. 3 Reactivity Measurement Results of Eu_2O_3-ZrO_2 or Eu_2O_3-HfO_2.

Table II Differences between Calculations and Measurements (C-E) of Reactivity for Eu_2O_3-ZrO_2 or Eu_2O_3-HfO_2

Eu_2O_3[g]	0	0.11	0.77	1.9	0	0.092	1.1	2.2	2.5
HfO_2 [g]	6.7	6.5	4.6	1.0	0	0	0	0	0
ZrO_2 [g]	0	0	0	0	6.6	6.6	4.6	1.1	0
C-E [pcm]	6.4	-	5.1	3.9	3.4	-1.4	-2.6	3.2	2.8

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