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 ^109Ag 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 ^235U, ^238U and ^239Pu are around 5eV. Therefore, it is confirmed that NRTA can be utilized for the measurements of nuclear fuel materials in the fuel pin sample.

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REFERENCES:
**INTRODUCTION:** In order to reduce TRU, the research and technology development entitled as “TRU burning fast reactor cycle using uranium-free TRU metallic 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 counter-measures [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 mm³ and 3.0 mm³ were prepared to identify the neutron self-shielding effects for each resonance. Those samples had cross sections of 2.0×2.0 cm². 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.

**RESULTS:** The measured TOF spectra with 1.0 mm³ and 3.0 mm³ 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 mm³ 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.

**REFERENCES:**

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 neutron 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 Bi4Ge3O12 (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 $7.36 \times 10^{-3}$ (lb) 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.

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:
CO2-4  Development of Gamma Imager and Its Application to Identification of Nuclear Materials


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2 Tokyo City University
3 National Institute of Technology, Toyama College
4 Institute for Integrated Radiation and Nuclear Science, Kyoto University

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 10B 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 10B 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 10B sample. Peaks corresponding to prompt gamma-ray of 10B (478 keV) and 112Cd (558, 651 keV etc.) appear clearly in the spectrum. Typical reconstructed image of 10B prompt gamma-ray on a projection sphere around the prototype CdTe detector is shown in Fig.3. Note that 10B 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 10B 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 112Cd 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 10B prompt gamma-ray used as a shielding material for thermal neutrons without any pro-}

![Fig. 1. Experimental setup at the pulsed neutron source.](image1)

![Fig. 2. Typical energy spectrum from the 10B sample.](image2)

![Fig. 3. Reconstructed images on the projection sphere (θ, φ), (a) with the 10B sample and (b) with a dummy sample not including 10B.](image3)

REFERENCES:
INTRODUCTION: Evaluation of neutron fluence and neutron dose equivalent for the epithermal 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 epithermal 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$(n,\alpha)$ reactions. We experimentally evaluate the neutron detection system for absolute measurements of epithermal neutrons. The epithermal 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$_{nat}$Gd$_{10}$B$_3$O$_9$:Ce$^+$ (LGB) scintillator was used as a boron containing scintillator. However, the LGB scintillator is not yet commonly used. In the present study, we tried to demonstrate epithermal 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]. Fig.1 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 scintillator. We are developing a data analysis algorithm. The experimental results will be compared with experimental results of the previous studies.

REFERENCES:
This work was supported by JSPS KAKENHI (JP16K21679, JP16K09030).
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:

$$^{40}\text{Ca}^{2+} (\text{aq}) + ^{48}\text{Ca}L^{2+} (\text{org}) \leftrightarrow ^{48}\text{Ca}^{2+} (\text{aq}) + ^{40}\text{Ca}L^{2+} (\text{org})$$ (1)

where L represents macrocyclic polyether(18-crown-6).

A 20 ml aqueous solution (3M CaCl₂) 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.

REFERENCES:

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$_2$O$_3$) are applied. The ATCR reduces the risk of re-criticality accident in any reactor conditions including severe accident (SA). For the ATCR candidate materials RE$_2$O$_3$-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. 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$_4$C or Ag-In-Cd alloy. In this study, the reactivity worths of rare-earth sesquioxides (RE$_2$O$_3$) are applied. The ATCR which the novel neutron absorbing materials including enhanced accident tolerant fuels and core components” are considered to be applicable as the alternative control materials.

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$_2$O$_3$-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 neutron activity worth of each sample in E3 core was calculated with 2.58 billion neutron histories. The statistical error of neutron activity worth of each sample in E3 core was calculated using Table I Differences between Calculations and Measurements (C-E) of Reactivity for Sm$_2$O$_3$-ZrO$_2$ or Sm$_2$O$_3$-HfO$_2$.

Table I Differences between Calculations and Measurements (C-E) of Reactivity for Sm$_2$O$_3$-ZrO$_2$ or Sm$_2$O$_3$-HfO$_2$.

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<th>RE$_2$O$_3$ [g]</th>
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<th>0.93</th>
<th>2.5</th>
<th>0</th>
<th>0.093</th>
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<td>C-E [pcm]</td>
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<td>8.6</td>
<td>5.8</td>
<td>2.9</td>
<td>3.4</td>
<td>3.2</td>
<td>-3.9</td>
<td>-2.3</td>
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Fig. 2 Measurement results of reactivity for Sm$_2$O$_3$-ZrO$_2$ or Sm$_2$O$_3$-HfO$_2$.

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<th>0.93</th>
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<th>0</th>
<th>0.093</th>
<th>1.3</th>
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<tr>
<td>C-E [pcm]</td>
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<td>7.0</td>
<td>5.6</td>
<td>3.1</td>
<td>0</td>
<td>0</td>
<td>6.6</td>
<td>6.6</td>
<td>4.6</td>
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</table>

Fig. 3 Reactivity Measurement Results of Eu$_2$O$_3$-ZrO$_2$ or Eu$_2$O$_3$-HfO$_2$.

Table II Differences between Calculations and Measurements (C-E) of Reactivity for Eu$_2$O$_3$-ZrO$_2$ or Eu$_2$O$_3$-HfO$_2$.

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<th>0.77</th>
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<tbody>
<tr>
<td>C-E [pcm]</td>
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<td>-</td>
<td>5.1</td>
<td>3.9</td>
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