CO2-1 High-Precision Mass Analysis of RI Sample for Cross-Section Measurements

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INTRODUCTION: The ImPACT project[1] aims to realize a "large reduction of and exploitation of resources in high-level nuclear waste by nuclear transmutation", by using an accelerator to accomplish nuclear transmutation of long-lived fission products (LLFPs). To perform accelerator-based nuclear transmutation effectively, it is necessary to acquire reaction cross-section data for radionuclides across a wide spectrum of incident particle energies. This project targets the major LLFPs: ^{107}Pd, ^{93}Zr, ^{135}Cs, ^{126}Sn, and ^{79}Se, as well as incidentally the medium-lived fission product ^{137}Cs. We targeted the ^{135}Cs because of its long half-life (2.3×10^{6} yr[2]). However, it is impossible to obtain a pure ^{135}Cs sample because it is not supplied by available vendors. This is also why the quantity of data is so scarce [3, 4]. Isotope separation of ^{135}Cs and ^{137}Cs is difficult, and therefore ^{135}Cs is included as an impurity in available ^{137}Cs samples. The isotope abundance of ^{135}Cs and ^{137}Cs in a standard ^{137}Cs source has been measured by mass spectrometry. Then, the amount of ^{135}Cs can be obtained from yields of 662 keV decay gamma-rays of ^{137}Cs and the isotope abundance. It is also necessary to suppress contamination of the mass spectrometer from radioactive ^{137}Cs and ^{135}Cs. It is necessary to perform highly precise analysis for an extremely small amount of sample. Therefore, this research aims to apply mass spectrometry to a very small amount of radioisotope sample, and to confirm its effectiveness.

EXPERIMENTS: A standard solution of ^{137}Cs (370kBq) was obtained through the Japan Radioisotope Association. An appropriate amount of the ^{137}Cs solution was dispensed, and then converted to nitrate by a chemical processing. The mass spectrometer TRITON was used for this analysis, which was made by Thermo Fisher Scientific. A small amount of the chemically processed solution, about 10Bq, was pipetted onto a Re filament with a TaO activator, and then dried. Three filaments with applied Cs solution were prepared. The filaments were attached onto the ion source of the mass spectrometer.

RESULTS AND DISCUSION: **Figure 1** shows the resulting mass spectrum of the standard ^{137}Cs sample. From the yields of the mass spectrum, the obtained ^{135}Cs/^{137}Cs isotope ratio was 0.868±0.004 (±2σ). Before the chemical processing, false peaks of about 1% were observed as the part of red line, which would come from sources other than Cs or molecular ion. The false peaks appear at Cs masses plus 0.1u. On the other hand, it is known that peaks from Ba appear on the almost same mass. Since the false peaks decayed faster than Cs peaks with the elapse of time, it is supposed that the false peaks were due to some kind of chemical impurities. In the case of the chemically processed sample, false peaks did not appear as shown with the black line in Fig.1. From this, it is thought that false peaks were due to some kind of chemical impurities in the Cs sample because the false peaks became extinct after chemical processing. The results of the isotope ratios in both cases were in agreement with each other within the limits of errors.

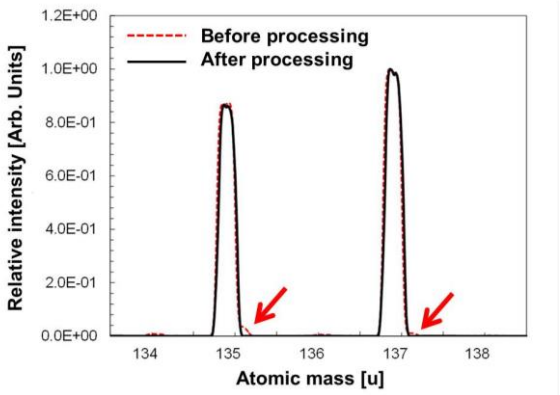

Fig. 1 Measured mass spectrum of ^{137}Cs sample

CONCLUSION: This work successfully measured the isotope ratio of ^{135}Cs and ^{137}Cs with an accuracy of 0.5% using a standard solution of ^{137}Cs. This technique makes it possible to analyze a very small amount (picogram order) of radioactive sample. When the Kyoto University Reactor is operated again in near future, neutron capture cross-section measurements of ^{135}Cs shall be performed by activation method with the ^{137}Cs solution analyzed in this work.

Acknowledgement

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CO2-2 Measurement of Doppler Effect by Small Accelerator Neutron Source (1)

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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. However, the differential experiment of the Doppler effects for Mo and Nb has not been carried out so far. Therefore, we have initiated the measurement of the Doppler effects for Mo sample by Time-of-flight (TOF) method with the KURRI-LINAC pulsed neutron source.

EXPERIMENTS: We measured the Doppler effects of Mo sample with the KURRI-LINAC pulsed-neutron source. In the experiment, neutron capture rates in Mo sample at 300 K and 600 K were obtained by prompt gamma-ray measurement with the TOF method. The Mo 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 natural molybdenum samples with different thickness of 0.5 mm^t and 3.0 mm^t were prepared to identify the neutron self-shielding effects for each resonance. Those were metallic plates of 2.0×2.0 cm^2. The measurements with thick and thin Mo samples, the graphite sample with 3.0 mm in thickness, and no sample (blank run) at 300 K and 600 K were carried out.

RESULTS: The measured TOF spectra with 0.5 mm^t and 3.0 mm^t Mo samples at 300K are shown in Fig. 1. The main resonances of Mo-95, Mo-96, Mo-97 and Mo-98 were clearly observed in the energy range from 10 eV to 1 keV. For further analysis, counting statistics observed in TOF spectrum were improved by rebinning the data into the JSF70 group structure [5] corresponding to the lethargy width of 0.25. Comparison of the rebinned TOF spectra for the 3.0 mm^t sample at 300 K and 600 K is shown in Fig. 1. The Doppler capture rate ratio defined as the capture rate at 600 K divided by it at 300 K was obtained for each energy group. The Doppler capture rate ratio were 1.3 ± 0.7 % at 44.9 eV, 8.0 ± 0.7 % at 70.9 eV, 1.0 ± 0.7% at 131.4 eV and 6.5 ± 3.0% at 358.6 eV resonances.

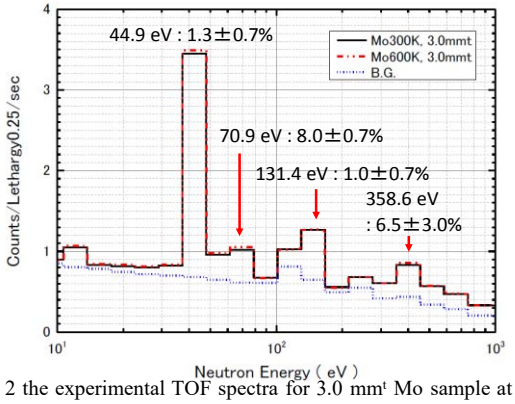

Fig. 2 the experimental TOF spectra for 3.0 mm^t Mo sample at 300 K and 600 K

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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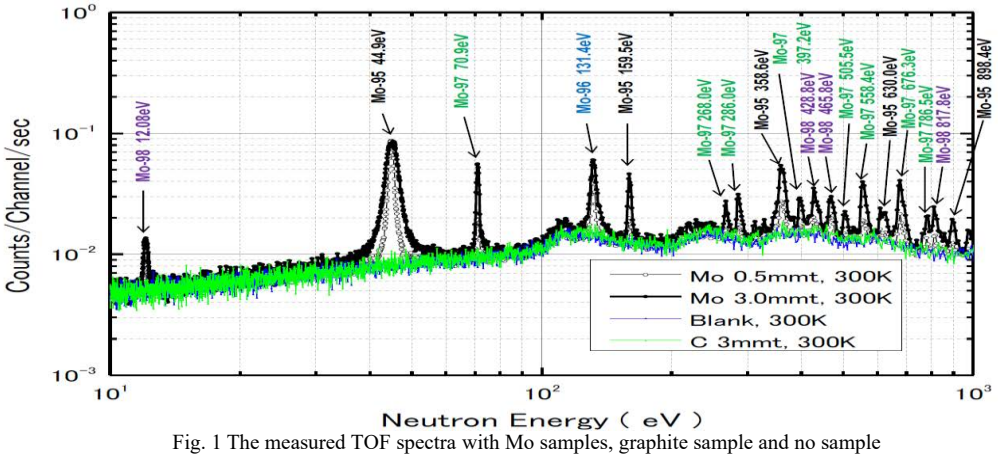

Fig. 1 The measured TOF spectra with Mo samples, graphite sample and no sample

CO2-3 Development of Neutron Detectors for Precise Measurement of Epi-thermal Neutrons

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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 (BNCT). It is not easy to determine precisely the neutron fluence for the epi-thermal neutrons in an irradiation field because of large uncertainty of reaction cross sections in the epi-thermal region. In the present study, we have developed neutron detectors for absolute measurement of epi-thermal neutrons, high intensity epi-thermal neutrons in a BNCT field and an imaging detector for a hidden material.

EXPERIMENTS: A collimated neutron beam was obtained by the photo-neutron reaction using a water-cooled tantalum target at the KURRI Linac [1]. We experimentally evaluated characteristics of the epi-thermal neutron detector for absolute measurement, the high intensity epi-thermal neutron detector and the imaging detector.

(1) Epi-thermal neutron detector for absolute measurement

The epi-thermal neutron detector for absolute measurement is composed of a $^{6}Li6^{nat}Gd^{10}B3O9$:Ce+ (LGB) scintillation detector and two BGO scintillation detectors. In our previous experiments, we used an NaI(Tl) scintillation detector instead of the BGO scintillation detectors. However, the NaI(Tl) scintillator has large sensitivity to neutrons as a comparison with the BGO scintillator. The 50 mm-diameter and 5-mm thick LGB scintillator was set at the center of the beam line. The 50.8 mm-diameter and 50.8 mm thick BGO scintillators were located on both sides of the LGB scintillator and out of neutron beam. When the LGB scintillator detects neutrons by the $^{10}B(n,\gamma\alpha)$ reaction, 478 keV monoenergetic gamma rays are produced and subsequently detected with the BGO scintillators. Moreover, the absolute neutron fluence is determined by measuring gamma rays from the $^{10}B(n,\gamma\alpha)$ reaction with the BGO scintillators in setting a 1-cm thick $^{10}B_4C$ total absorption sample in front of the LGB scintillator. Number of alpha particles and gamma rays produced by the $^{10}B(n,\alpha\gamma)$ reaction in the LGB scintillator are absolutely determined by the coincidence measurements. Pulse height spectra of the LGB and BGO scintillators from the $^{10}B(n,\alpha\gamma)$ reaction were obtained in the coincidence measurements. However, the evaluation of the background due to scattered neutrons was not sufficient. In future, we will improve the measurement process based on the present experimental results.

(2) High intensity epi-thermal neutron detector

The high intensity epi-thermal neutron detector is important in the BNCT. The detector is simply composed of a ^{6}Li-glass scintillator and a ^{7}Li-glass scintillator and photomultipliers (PMTs). Current from an anode output of the PMT is detected by a current integrator. The ^{7}Li-glass scintillator is used to subtract the gamma ray contribution detected by the ^{6}Li-glass scintillator. We experimentally evaluated the difference between the neutron detection efficiencies of the ^{6}Li-glass and ^{7}Li-glass scintillators using the Linac neutron source. Finally, we successfully obtained the relation between the thermal neutron flux and output current using the neutron detection efficiencies in the thermal neutron standard field of the National Institute of Advanced Industrial Science and Technology. In future, we will perform the high flux neutron irradiation using the KUR.

(3) Imaging detector

The imaging detector is used to search hidden materials such as nuclear materials and radioactive sources. The imaging detector is composed of multi-pixel type CdTe detectors. The imaging detector detects prompt gamma rays produced by the neutron capture reaction in the hidden materials or gamma rays from a hidden radioactive source. After that, the imaging detector determines a position of the hidden material or the hidden radioactive source. Characteristics of the detector were evaluated using gamma rays from the neutron capture reactions of ^{10}B and ^{197}Au. We will continue the experiments in order to verify potential of the practical realization using the TOF method.

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CO2-4 Research and Development for Accracy Improvement of Neutron Nuclear Data on Long-lived Radioactive Nnuclei at KURRI-Linac

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INTRODUCTION: There are strong requests for reducing the uncertainty of neutron capture cross section data of minor actinides (MAs) to estimate the transmutation rate of those long-lived radioactive nuclei in the innovative reactor system. In recent years, intense pulsed spallation neutron sources became available to remarkably improve the precision of neutron TOF data. However, there are discrepancies out of a range of tolerance between current experimental results. It is understood that the unrecognized systematic errors make a difference. In order to recognize and reduce the systematic errors, the project entitled as "Research and development for Accuracy Improvement of neutron nuclear data on Minor ACtinides (AIMAC)" has been started [1]. In this project, we aim at obtaining the resonance parameters precisely of MAs by combining the neutron capture γ-ray measurement to transmission neutron measurement at KURRI-Linac. Neptunium-237 is one of the most important MAs with a long half-life. In this year, neutron total cross section and capture cross section of ^{237}Np were measured and resonance analysis was also performed.

EXPERIMENTS: The total and capture cross section measurements have been performed by the neutron time-of-flight (TOF) method using the 46-MeV electron linear accelerator at the Research Reactor Institute, Kyoto University (KURRI-LINAC). Transmitted neutrons were detected by a 6 mm thick GS20 ^{6}Li-glass scintillator. Neptunium oxide powder of 1.13 g packed in an aluminum disk container of 30mm in diameter and 0.4 mm thick wall, which was placed at a distance of 10.15 m from the photo-neutron source. An aluminum disk container without the neptunium oxide powder was also used as a dummy case. The linac was operated with an electron energy of about 30 MeV, an averaged beam current of 17 μA, a repetition rate of 50 Hz, a pulse width of 0.1 μs. Neutron capture gamma rays from the sample were measured with a 4π bismuth germinate (BGO) scintillation detectors composed of 12 BGO cylindrical crystals having 2 inch. in diameter and 2 inch. in length. A capture sample was set in the center of the detector. Three samples of ^{237}Np with different thickness were used; the activities of the samples were 26, 5.2 and 1 MBq, respectively. The incident neutron flux shape was measured with a ^{10}B sample.

RESULTS: Preliminary result of total cross section of ^{237}Np is shown in Fig. 1. The resonance analysis of the deduced cross section values were performed with the SAMMY code [2] in the energy range from 10^{-2} to 80 eV. The resonance parameters of the 0.49-eV resonance were modified to reproduce the present results. The neutron capture yields for the samples with different thickness were also obtained as shown in Fig. 2. The neutron self-shielding and multiple-scattering effect in the sample depends on the resonance parameter. Therefore, the data were used for the cross-check of resonance parameters.

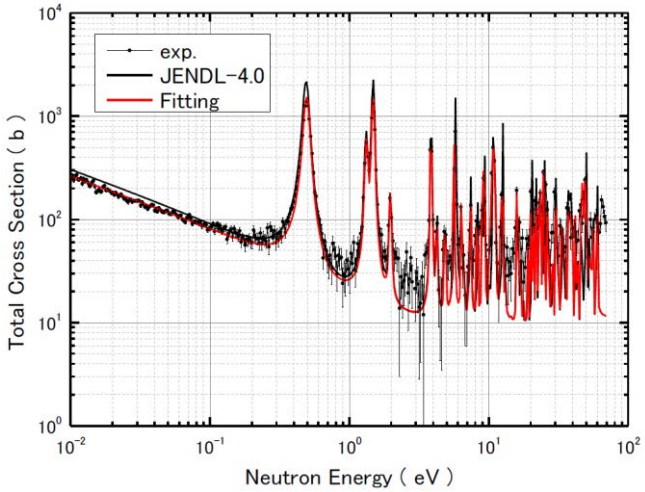

Fig. 1 Preliminary results of neutron total cross section of ^{237}Np

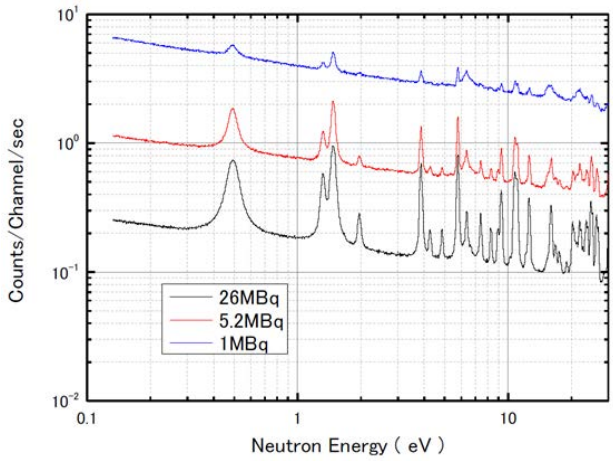

Fig. 2 Capture yields for three ^{237}Np samples measured by the BGO spectrometer

Present study includes the result of "Research and Development for accuracy improvement of neutron nuclear data on minor actinides" entrusted to the Japan Atomic Energy Agency by the Ministry of Education, Culture, Sports, Science and Technology of Japan (MEXT).

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