Neutron Capture Cross Section Measurement of Minor Actinides in Fast Neutron Energy Region for Study on Nuclear Transmutation System

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A research project entitled *"Study on accuracy improvement of fast-neutron capture reaction data of long-lived MAs for development of nuclear transmutation systems"* started in 2017 as a four-year project. The purpose of the project is to improve the neutron capture cross sections of minor actinides in the fast neutron energy region that is particularly important for study on a nuclear transmutation system. The outline of the project and the current progress are reported.

1. Introduction

Disposal of high-level nuclear waste from nuclear power plants has been a long-standing issue in the nuclear industries. Nuclear waste contains long-lived minor actinides (MAs), some of which keep their radiotoxicities for more than thousand years. Currently planned geological disposal of high-level nuclear waste has been in deadlock due to the difficulty of public acceptance for many years. In order to solve this issue, nuclear transmutation, by which long-lived MAs are converted to stable or shorter-lived nuclides via neutron-induced reactions, has been suggested. In recent years, accelerator-driven systems (ADS) are considered as feasible candidates of MA burners and several ADS projects are ongoing or planned. Detailed core design of an ADS requires accurate, reliable nuclear reaction data of MAs but the uncertainties of the current cross section data in evaluated nuclear data libraries in the fast neutron energy region that is most relevant for ADS are not small enough to satisfy the requirement [1,2].

A research project entitled *"Study on accuracy improvement of fast-neutron capture reaction data of long-lived MAs for development of nuclear transmutation systems"* started in 2017. The project aims at improving the accuracies of neutron capture cross sections of MAs (^{237}Np, ^{241}Am, ^{243}Am) in the fast neutron energy region. In order to improve the capture reaction data of MAs, an intense pulsed neutron beam from a spallation neutron source of the Japan Proton Accelerator Research Complex (J-PARC) is utilized in time-of-flight (TOF) experiments to measure capture cross sections. In previous research projects, we worked on building and commissioning the Accurate Neutron Nucleus Reaction Measurement Instrument (ANNRI), a neutron beam line for nuclear data measurement in the Materials and Life Science Experimental Facility (MLF) of J-PARC [3]. ANNRI now becomes one of the leading neutron beam lines for nuclear data measurement in the world.

One of the major reasons why previous measurements of MA capture cross sections were not able to achieve high accuracies is that MA samples were radioactive. A radioactive sample emits decay γ-rays that become background for the detection of neutron capture γ-rays. The large decay γ-ray background hinders accurate capture γ-ray measurement. The J-PARC high-intensity pulsed neutron beam solves the issue by increasing neutron capture reaction rates in a sample. The rate of capture events to background events can be improved, consequently achieving small uncertainties of cross sections.

This project consists of four tasks: (1) development of neutron beam filter system in J-PARC, (2) neutron capture cross section measurement, (3) sample characteristic assay, and (4) theoretical reaction model study. The following sections describe details of the tasks.

2. Neutron Beam Filter System

The neutron beam filter is designed to solve the so-called double bunch issue of a neutron beam from the J-PARC spallation neutron source. The spallation neutron source is operated at a repetition rate of 25 Hz. Hence, neutrons are generated every 40 ms. The J-PARC accelerator adopts a special operational pattern called double bunch operation, in which two proton beam pulses with a separation time of 600 ns are injected into the spallation neutron target for each neutron burst cycle. The purpose of this operation is to increase the thermal neutron intensity, important for most of measurements with the neutron beam lines in MLF. In the thermal neutron TOF range, the time structure of the incident proton beam is negligible. The Doppler broadening effect and moderation time in a moderator erase the proton beam time structure in the thermal neutron TOF region over 12 ms. On the other hand, this double bunch mode is very problematic for measurements in the high energy region above 100 eV, where the double bunch structure appears in TOF spectra. Neutrons having two different energies originating from two incident proton pulses overlap at the same TOF in measurement. The capture yield at each TOF point is contributed by two different neutron energies and the results cannot be deconvoluted easily.

A neutron beam filter system solves this issue. The neutron beam filter method is often used in nuclear reactor experiments [4,5]. A reactor neutron beam which has a continuous energy distribution can be tailored to be mono or quasi-mono energetic through filter materials that have sharp resonance dips of total cross section at certain resonance energies. In this project, the neutron filter technique is combined with the TOF technique to separate out coexisting different energy neutrons at the same TOF. The neutron beam filter system is under development. Filter materials Fe, Bi, Al, Si, Cr and Sc were chosen and tested in 2018. Based on the test results, the system was designed and installed in the ANNRI beam line of J-PARC MLF. A measured neutron energy spectrum filtered with Fe is shown in Fig. 1. A neutron spectrum with no filter is also shown for comparison. As seen in Fig. 1, a prominent peak is clearly observed at an energy of 24 keV.

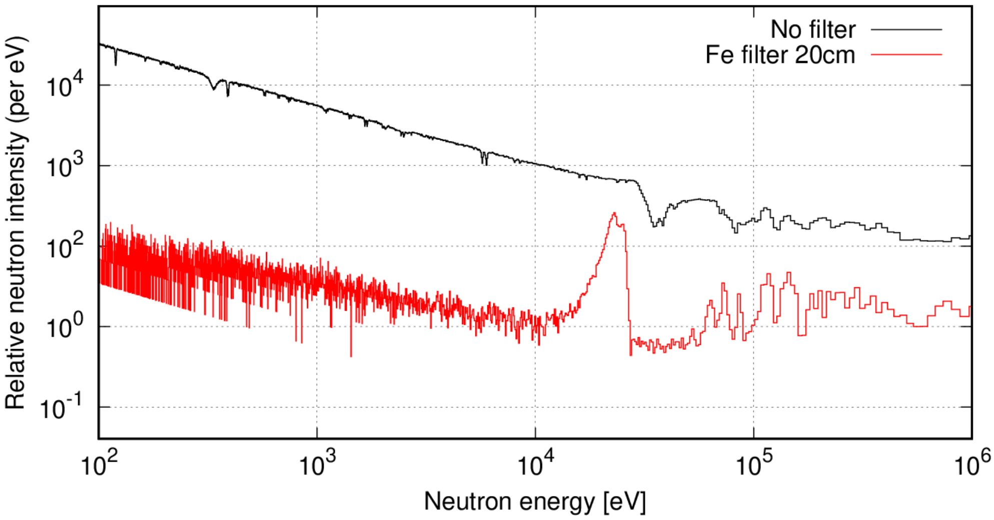

Fig. 1 Neutron energy spectrum filtered with Fe (blue). A neutron spectrum with no filter (red) is shown for comparison.

3. Neutron Capture Cross Section Measurement

This project focuses on fast-neutron capture cross section data of MA. This requires a fast detection and data acquisition systems. Fast neutron events appear in fast TOF region close to the gamma flash, an intense γ-ray emission produced at the moment that the incident proton beam pulse reaches the spallation neutron target. The gamma flash overwhelms the detection system and detection signals are distorted for μs (sometimes ms) after the gamma flash. To detect neutron capture events in the fast TOF region, the system needs to recover quickly from the distortion caused by the gamma flash. In addition to the gamma-flash, an intense neutron beam from the J-PARC spallation neutron source increase the detector counting rate, leading to large count loss due to the system dead time.

We plan to measure the neutron capture cross sections of MAs using NaI(Tl) detectors of ANNRI. NaI(Tl) detectors are suitable for the measurement in fast TOF region [6]. Scintillation detectors have faster response than semiconductor detectors, and what's more, an NaI(Tl) detector can measure a γ-ray spectrum. The pulse height weighting technique to derive neutron capture cross sections is well established for a NaI(Tl) detector [7].

In addition, we developed a new method to determine the absolute neutron capture yields using a sample rotation system. The effective sample thickness can be changed by tilting the sample with respect to the beam axis. The self-shielding factor changes with the effective sample thickness. The absolute neutron capture yield is determined from the self-shielding factor that is calculated from change of capture yield by tilting the sample.

4. Sample Characteristic Assay

Sample characteristic assay is an important task to improve the accuracy of cross sections. Uncertainties of sample characteristics such as total mass, isotope composition and impurities can be crucial systematic uncertainties. In this project, we plan to analyze the isotope composition and impurities of MA samples by thermal ionization mass spectrometry (TIMS) at the Institute of Integrated Radiation and Nuclear Science of Kyoto University. The target accuracy of analysis is set at less than 1% in this project. The key to achieving such a high accuracy is the stability of ion emission from a filament of the TIMS ion source. To stabilize the ion emission, the monitoring method of the filament temperature, most dominant factor for ion emission, was improved in 2018. A test experiment showed significant improvement of ion emission control. TIMS analysis of MA samples is planned to conduct in 2020.

5. Theoretical Reaction Model Study

Theoretical nuclear reaction models can predict neutron capture cross sections. Combined with experimental data, theoretical reaction models become powerful tools in nuclear data evaluation. In this project, neutron capture γ-ray spectra measured with the ANNRI-NaI(Tl) detectors are used to refine theoretical model prediction. Capture γ-ray spectra give more information on reaction mechanisms than only from capture cross section. Comparing theoretical calculations with the measured γ-ray spectra, model parameters such as gamma-ray strength function and level density can be determined. However, direct output spectra from experiments cannot be compared with theoretical capture γ-ray spectra because measured spectra convolute detector response. Unfolding measured spectra with detector response function is often performed to compare with theoretical spectra [7] but the unfolding process adds uncertainties to experimental data. Instead, we adopted the reverse process for comparison. We fold theoretical spectra with detector response and then compare them to experimental data. Folding process is less ambiguous than unfolding process. We built a geometrical model of the ANNRI-NaI(Tl) detectors for the Monte Carlo simulation code PHITS [8] and calculated the detector response matrix. For a benchmark calculation, the capture γ-ray spectrum of ^{197}Au was calculated with the theoretical reaction model code CCONE [9] and then folded the calculated spectrum with the detector response matrix

6. Summary and Future Prospect

The project entitled *"Study on accuracy improvement of fast-neutron capture reaction data of long-lived MAs for development of nuclear transmutation systems"* started in 2017 as a four-year project. The first two years were spent for development of the neutron beam filter system, fast data acquisition method for the ANNRI-NaI(Tl) detectors and MA sample characteristic assay. The neutron filter system and the sample rotation system were installed in ANNRI. Now the project is in the actual experimental phase to measure the capture cross sections and capture γ-ray spectra of MAs.

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References

[1] M. Salvatores *et al.*, OECD/NEA Report No.6410 (2008).
[2] H. Iwamoto *et al.*, J. Nucl. Sci. Technol., **50**, 856 (2013).
[3] K. Kino *et al.*, Nucl. Instr. Meth. A, **736**, 66 (2014).
[4] O. D. Simpson and L. G. Miller, Nucl. Instr. Meth, **61**, 245 (1968).
[5] A. T. Tran *et al.*, Ann. Nucl. En. **113**, 420 (2018).
[6] T. Katabuchi *et al.*, Nucl. Instr. Meth. A, **764**, 369 (2014).
[7] S. Mizuno *et al.*, J. Nucl. Sci Technol., **36**, 493 (1999).
[8] T. Sato *et al.*, J. Nucl. Sci Technol., **55**, 684 (2018).
[9] O. Iwamoto *et al.*, Nucl. Data Sheets, **131**, 259 (2016).