Experimental program of nuclear data for accelerator-driven nuclear transmutation system using FFAG accelerator –First subprogram: spallation neutron measurement

Keita NAKANO^{†1}, Hiroki IWAMOTO^{1,2}, Shin-ichiro MEIGO¹, Katsuhisa NISHIO³, Yoshihiro ISHI⁴, Kentaro HIROSE³, Yosuke IWAMOTO², Yasutoshi KURIYAMA⁴, Fujio MAEKAWA¹, Hiroyuki MAKII³, Kota OKABE¹, Riccardo ORLANDI³, Akito OIZUMI², Daiki SATOH², Fumi SUZAKI³, Kazuaki TSUKADA³, Tomonori UESUGI⁴, Hiroshi YASHIMA⁴, and Yoshiharu MORI⁴

¹J-PARC Center, Japan Atomic Energy Agency, Tokai-mura, Ibaraki 319-1195, Japan ²Nuclear Science and Engineering Center, Japan Atomic Energy Agency, Tokai-mura, Ibaraki 319-1195, Japan

³Advanced Science Research Center, Japan Atomic Energy Agency, Tokai-mura, Ibaraki 319-1195, Japan

⁴Institute for Integrated Radiation and Nuclear Science, Kyoto University, Kumatori-cho, Osaka 590-0494, Japan [†]Email: nakano.keita@jaea.go.jp

Abstract

For accurate prediction of neutronic characteristics for accelerator-driven nuclear transmutation system (ADS) and a source term of spallation neutrons for reactor physics experiments for the ADS at Kyoto University Critical Assembly (KUCA), we have launched an experimental program to measure nuclear data on ADS using the Fixed Field Alternating Gradient (FFAG) accelerator at Kyoto University (Period: October 2019 – March 2023). This program consists of two subprograms, focusing on two nuclear reaction mechanisms, (1) spallation reactions and (2) high-energy fission, for incident proton energies from several tens of MeV to 100 MeV. In the first subprogram, we will measure double-differential cross-sections (DDXs) and thick-target neutron-yields (TTNYs) for several targets (*i.e.*, Pb, Bi, and Fe); in the second subprogram, mass distribution of fission product and neutron multiplicity in high energy fission of Pb and Bi will be measured. In this paper, the present status of the first subprogram is described.

1 Introduction

Accelerator-driven nuclear transmutation systems (ADSs) have attracted attention as one of the promising options to solve the nuclear issue of high-level radioactive waste management [1]. In terms of the neutronic design of the ADS, precise experimental nuclear data and reliable theoretical nuclear models are required. So far, much effort has been devoted to measure nuclear data at incident proton energies from several hundred MeV to GeV range, and these experimental nuclear data play a significant role in the improvement of the nuclear models. However, it has been pointed out that lack of nuclear data in the energy range from several tens of MeV to



Figure 1: Schematic drawing of a vacuum chamber and neutron detectors.

100 MeV makes it difficult to improve the nuclear models, which results in large uncertainty in the neutronic characteristics of the ADS. Satoh *et al.* demonstrated that recent nuclear models such as the Liège Intranuclear Cascade model [2] failed to predict the measured spectra of emitted neutrons in the most-forward direction at the incident proton energy below 78 MeV [3]. Therefore, accumulation of experimental data on important nuclides in ADS such as neutron production cross sections in proton-induced reactions at incident energies from several tens of MeV to 100 MeV and mass distribution of fission product and neutron multiplicity from high energy fission are strongly desired.

In this circumstance, a new experimental campaign has been launched to measure nuclear data related to ADS. The campaign consists of two subprograms: measurement of neutron energy spectra from spallation reactions of Fe, Pb, and Bi by protons from several tens of MeV to 100 MeV and that of mass distribution of fission product and fission multiplicity in high energy fission of Pb and Bi. For the first subprogram, the Double-Differential neutron production cross sections (DDXs) and Thick-Target Neutron Yields (TTNYs) for such materials will be measured using a proton beam accelerated by Fixed Field Alternating Gradient (FFAG) accelerator at Kyoto University. In this paper, the experimental plan of the first subprogram and a detector test performed for the experiment are described.

2 Plan of Experiment at FFAG Facility

The experiment of the first subprogram will be conducted at FFAG Facility, Kyoto University. In this experiment, proton-induced DDXs and TTNYs for Fe, Pb, and Bi targets will be measured using the time-of-flight method. Also, sensitivity check of a ²³⁷Np fission chamber under the spallation neutron field will be investigated. Figure 1 shows a photo of the FFAG facility and a schematic drawing of a vacuum chamber and neutron detectors. One of the targets is placed at the center of the chamber. Target thicknesses for the DDX experiment are 2 mm for all targets to achieve enough statistics, and those for the TTNY experiment are 30 mm for the Fe



Figure 2: Configuration of the measurement geometry at FRS.

and Pb targets and 35 mm for the Bi target, which are designed so that 150-MeV protons stop in the targets. The range of incident protons was calculated by SRIM code [4]. Diameters of the targets for both the DDX and TTNY experiments are 48 mm which is larger than beam size of 40 mm calculated by beam optics. An NE213 liquid organic scintillator (size: 20 mm in length and 8 mm in diameter) coupled with a photomultiplier tube (H3164, Hamamatsu Photonics) will be placed 5 m far from the target to detect neutrons. Signals from the detector will be fed to a digitizer (SIS3316). Current of the proton beam is planed to be 0.1 nA for the TTNY measurement and 1 nA for the DDX measurement. To eliminate contribution of roomback neutrons to the DDX and TTNY experiments, room-back neutron measurements will also be conducted with a shadow bar made of stainless steel (size: 1 m in length and 50 mm in diameter), whose length is designed to reduce the neutrons produced from the targets to several percent. Measurements at several emission angles will be conducted by changing the position of the detector. The ²³⁷Np fission chamber (size: 25.4 mm in length and 6 mm in diameter) will be used to check the reaction rate under the spallation neutron field around the target vacuum chamber. The activity of coated ²³⁷Np is 31.3 kBq on June 30, 1982.

3 Detector test at Facility of Radiation Standard

In advance of the experiment at the FFAG facility, measurement of neutron detection efficiency for the neutron detector (NE213 + H3164) and response of the ²³⁷Np fission chamber were conducted at Facility of Radiation Standard (FRS) [5], Japan Atomic Energy Agency (JAEA). In this experiment, 14.8 MeV monoenergetic neutrons generated by T(d, n) reaction was used. The RF repetition frequency was 4 MHz. Figure 2 shows a photo of the measurement geometry. Neutrons generated at a production target directly irradiated to the neutron detectors such as the NE213 and the ²³⁷Np fission chamber placed at approximately 1 m downstream to the beam port at 60 degree from the most-forward direction. The measurement with a shadow bar was also performed in the NE213 measurement to estimate background neutrons. Scintillation photons produced in the NE213 were converted to electric pulses by the photomultiplier tube and fed to the digitizer after passing through a 40 meter-long low-attenuation cable. To determine the time-of-flight, RF signals of the accelerator were used as start timing and the signals from NE213 as stop timing. The ²³⁷Np fission chamber was connected to a preamplifier and a



Figure 3: Two-dimensional plot of the obtained signal by the time-of-flight and the charge spectrum for the NE213 before (Left) and after (Right) the gamma-ray elimination.

spectroscopy amplifier. Then signals from the spectroscopy amplifier were fed to the digitizer and a multichannel analyzer.

4 Results and Discussion

4.1 Result for NE213 Detector

Figure 3 shows two-dimensional plot by the time-of-flight and the charge spectrum for the NE213 neutron detector. Three peaks at approximately 15, 30, and 50 ns can be seen, which correspond to prompt gamma-rays, 14.8-MeV neutrons, and 3.0-MeV parasitic neutrons, respectively. The parasitic neutrons were produced by the D(d, n) reactions between incident deuteron beam and deuterium accumulated in the tritium target. The neutron energies were estimated from the flight length and the difference of time-of-flight between the prompt gamma-rays and the neutrons. The gamma-ray events were rejected by the two-gate method of the pulse shape analysis. After eliminating the gamma-ray events, the number of detected 14.8-MeV neutrons is estimated with paying attention to the parasitic neutrons. The detection efficiency of the NE213 neutron detector was calculated from the neutron fluence measured by a reference long counter consists of a thermal neutron detector and a polyethylene moderator and the obtained counts by the NE213 neutron detector. The uncertainty of the neutron fluence measured by the long counter was 6%. Figure 4 shows the detection efficiency of the NE213 neutron detector as a function of the neutron energy. The black circles correspond to the ²⁴¹Am-quarter and ¹³⁷Cs-double biased efficiencies measured in the present experiment. Here the ²⁴¹Am-quarter biased efficiency means to neutron detection efficiency with larger light output than quarter of 59-keV electron equivalent (keVee) which corresponds to a photo peak of 59-keV γ -ray from ²⁴¹Am. Also the ¹³⁷Cs-double biased efficiency corresponds to that with larger output than 986 keVee [6]. The black square is the ²⁴¹Am-quarter biased efficiency measured in the previous experiment at lower neutron energy [7]. The solid and dashed lines are calculated efficiencies by SCINFUL-R [8, 9] code in each bias. The calculations almost reproduce the obtained data. However, small difference can be seen in the ²⁴¹Am-quarter biased efficiency. It is considered that the parasitic neutrons is not completely removed in the time-of-flight spectrum. Time



Figure 4: Detection efficiency of NE213 as a function of neutron energy [7].

resolution of the time-of-flight is approximately 4 ns which is originated from the inner clock of the digitizer, the pulse width of incident deuteron beam, and the flight path length.

4.2 Result for ²³⁷Np fission chamber

Figure 5 shows charge distribution from the ²³⁷Np fission chamber. The large number of low charge events and several high charge events are seen. The former is signals induced by α -decay of ²³⁷Np nuclei and the latter are fission events of them. The fission events are clearly separated from the α -decay component by their much larger deposit energy. We assume that the events with charge larger than 4.5×10^4 channel are fission events. The counts per fluence is estimated to be 9.0 ± 2.5 counts/(n/cm²). We also derive the calculated one from fission cross section of ²³⁷Np at 14.8-MeV neutron and the number of ²³⁷Np nuclei calculated by the activity of deposited ²³⁷Np. The calculated one is 6.8 counts/(n/cm²), which agrees with the measured one within 1σ of the statistical error. However, the statistics is little bit poor to check the consistency between the measured count rate and the calculated one. Also, the contribution of the parasitic neutron as seen in the time-of-flight spectrum measured by the NE213 should be considered carefully because ²³⁷Np has a comparable fission cross section at 3-MeV neutron.

5 Summary

For the development and design of ADS, more nuclear data measurements are desired especially the proton incident energy ranging from several tens of MeV to 100 MeV. In this situation, new experimental campaign has been launched to measure the TTNYs and the DDXs at FFAG facility, Kyoto University, and mass distribution of fission product and neutron multiplicity in high energy fission. At present, design of the measurement and detector tests are in progress. In advance of the experiment at FFAG, the detectors planned to be used in FFAG facility were tested with 14.8-MeV monoenergetic neutron at FRS, JAEA. The detection efficiency of NE213 liquid organic scintillator shows good agreement with calculation by SCINFUL-R. Also, count rate of ²³⁷Np fission chamber under the 14.8-MeV neutron field was consistent with the calculated value from the activity of deposited ²³⁷Np. From these measurements, the detector



Figure 5: The measured charge distribution from 237 Np fission chamber.

test and response check have been done and tuning towards the FFAG experiment will be conducted.

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