Measurement of neutron capture gamma rays from the resonances of ⁹³Zr at the J-PARC/MLF/ANNRI

J. Hori^{a*}, T. Fujii^a, S. Fukutani^a, M. Furusaka^d, K. Furutaka^b, S. Goko^{b,1}, H. Harada^b, F. Hiraga^d, M. Igashira^c, T. Kamiyama^d, T. Katabuchi^c, A. Kimura^b, T. Kin^b, K. Kino^{d,2}, F. Kitatani^b, Y. Kiyanagi^d, M. Koizumi^b, M. Mizumoto^c, S. Nakamura^b, M. Ohta^{b,3}, M. Oshima^b, K. Takamiya^a, Y. Toh^b

^aResearch Reactor Institute, Kyoto University, 2-1010, Kumatori-cho, Sennan-gun, Osaka, 590-0494, Japan

^bNuclear Science and Engineering Directorate, Japan Atomic Energy Agency, Tokai-mura, Naka-gun, Ibaraki, 319-1195, Japan

^c Research Laboratory for Nuclear Reactors, Tokyo Institute of Technology, O-okayama, Meguro-ku, Tokyo, 152-8550, Japan

^d Graduate School of Engineering, Hokkaido University, Kita-ku, Sapporo, Hokkaido, 060-8628, Japan Present address: ¹ Hokkaido University, ² High Energy Accelerator Research Organization, ³ Osaka University *Corresponding author: hori@rri.kyoto-u.ac.jp

1. Introduction

Neutron capture cross section data of minor actinides (MAs) and long-lived fission products (LLFPs) are required for design of advanced nuclear reactors such as fast reactors and accelerator driven systems. However, the current status of experimental data is not sufficient both in quality and in quantity. This is mainly because it is not easy to prepare enough amount of samples with high purity. To overcome the difficulty, we have started a series of experimental studies for MAs and LLFPs Accurate Neutron-Nucleus using the Reaction measurement Instrument (ANNRI) which was installed at the neutron Bean Line No.4 (BL04) of the Material and Life science experimental Facility (MLF) in the Japan Proton Accelerator Research Complex (J-PARC) [1]. The highly intense neutron source makes it possible to measure the cross sections accurately with a small amount of sample.

Though ⁹³Zr is one of the most important LLFPs, the isotopic purity of a sample is especially poor. Therefore, it is essential to separate the true capture events from the background due to impurities. A promising way is to measure discrete gamma rays with a high energy resolution and distinguish the true capture events. In this presentation, we report the relative intensities of neutron capture gamma rays emitted from the resonances of ⁹³Zr, which give an important information to apply the technique to the measurement of capture cross sections.

2. Methods and Results

2.1 Sample

The ⁹³Zr sample used in the present experiment was purchased from the Oak Ridge National Laboratory (ORNL). The zirconium oxide powder of 3.31 g was packed at the central region, 20 mm in diameter and 5.3 mm thick, of an aluminum disk-shaped container 30 mm in diameter and 6.3 mm thick. The isotopic purity of ⁹³Zr was determined with a Thermal Ionization Mass Spectrometer (TIMS) as 18.9 %. The stable isotopes of

Zr and the radioactive impurity of 125 Sb are also contained in the sample.

A NaCl crystal of 0.50 g was also used as a standard sample. The full-energy-peak efficiencies of the spectrometer were determined using well-known prompt gamma rays from the ${}^{35}\text{Cl}(n_{\text{th}},\gamma)$ reaction.

2.2 TOF facility and $4\pi Ge$ spectrometer

Pulsed neutron beam was produced by spallation reaction with 3-GeV proton beam bombarded on the mercury target at Japan Spallation Neutron Source (JSNS) [2]. In the measurement, MLF was operated at an average beam power of 120 kW and a repetition rate of 25 Hz in doubly bunched mode. The time interval between double pulses was 600 ns. The moderated neutrons were led to the BL04 and collimated to a diameter of 3 mm at a sample position using a rotary collimator system [3].

The capture gamma rays emitted from the sample were measured with the " 4π Ge spectrometer" which is composed of two cluster Ge detectors and eight coaxial Ge detectors surrounded by BGO anti-coincidence shields [4]. Each sample was set at the center of the spectrometer. The distance between the neutron source and the sample is 21.5 m. The signals from the Ge crystals were stored using a data acquisition (DAQ) system [5] in event-by-event mode as gamma-ray pulse-height and TOF data.

2.3 TOF spectrum

The TOF spectrum obtained by measuring the prompt gamma rays from the ⁹³Zr sample for 52 hours is shown in **Fig. 1**. The time-of-flight channel in a horizontal axis is converted into the neutron energy. In the spectrum, we can observe clearly two resonances of ⁹³Zr at 110 and 225 eV. The resonances of ⁹¹Zr and ⁹⁶Zr contained in the sample as isotopic impurities were also observed at 182, 240, 292 and 301 eV. The resonances above 150 eV are split into two peaks due to the doubly-bunched-beam effect. Two gates were set in the TOF spectrum to



Fig. 1 A TOF spectrum obtained for ⁹³Zr sample

obtain the pulse-height spectra corresponding to 110and 225-eV resonances as shown in Fig. 1.

2.4 Gamma-ray pulse-height spectra

Gamma-ray pulse-height spectra obtained by applying gates on the TOF regions for the 110- and 225-eV resonances are shown in **Fig. 2**. The very strong ground-state transitions at 919 and 1671 keV are remarkable. The true capture events can be distinguished effectively by using those ground-state transitions.

The number of assigned primary gamma-ray transitions were 9 and 7 for the 110- and 225-eV resonances, respectively. The intensity of each gamma-ray transition was obtained by dividing the net count of corresponding full-energy peak by the full-energy-peak efficiency of the spectrometer. **Figure 3** shows the relative intensity of each primary transition. It is found that the decay pattern is quite different between two resonances. It is worth noting that the primary transitions from the p-wave 225-eV resonance state to the low-lying states of 1-, 2+, 4+ were clearly observed. The spin and parity of the 225-eV resonance was



Fig. 2 Gamma-ray pulse-height spectra for the 110- and 225eV resonances of 93 Zr. The energies of primary gamma-ray peak with symbols are a: 8217 keV, b: 7298 keV, c: 6543 keV, d: 6161 keV, e: 6067 keV, f: 5887 keV, g: 5851 keV, h: 5709 keV, i: 5528 keV, j: 5372 keV, k: 4808 keV, l: 4494 keV, m: 4441 keV, respectively. Single-escape and doubleescape peaks are indicated by the arrows with s.e. and d.e., respectively.



Fig. 3 The relative intensities of primary transitions for the 110- and 225-eV resonances of ⁹³Zr. The energy, spin and parity of each final state are also indicated.

reported as 4- [6]. Assuming the E1 or E2 transition for the observed gamma rays, the spin of the 225-eV resonance is assigned to 3.

3. Conclusions

We have performed the measurement of neutron capture gamma rays from the 110- and 225- eV resonances of ⁹³Zr with the 4π Ge spectrometer at the ANNRI/MLF/J-PARC. The relative intensities of the observed primary transitions were derived. The spin of the 225-eV resonance of ⁹³Zr is assigned to 3. The present result shows that the 4π Ge spectrometer installed at the J-PARC/MLF/ANNRI is a unique tool for prompt gamma-ray analysis and J^{π} assignment of resonances.

Present study includes the result of "Study on nuclear data by using a high intensity pulsed neutron source for advanced nuclear system" entrusted to Hokkaido University by the Ministry of Education, Culture, Sports, Science and Technology of Japan (MEXT).

REFERENCES

[1] M. Igashira, Y. Kiyanagi, M. Oshima, "Nuclear data study at J-PARC BL04," *Nucl. Instrum. Methods A*, **600**, 332 (2009).

[2] Y. Ikeda, "J-PARC status update," *Nucl. Instrum. Methods A*, **600**, 1 (2009).

[3] K. Kino, *et al.*, "Measurement of energy spectra and special distributions of neutron beams provided by the ANNRI beamline for capture cross-section measurements at the J-PARC/MLF," *Nucl. Instrum. Methods A*, **626-627**, 58 (2011).

[4] T. Kin, *et al.*, "Development of a 4π Germanium Spectrometer for Nuclear Data Measurements at J-PARC," the 2009 NSS-MIC Conf. Rec., N24-2 (2009).

[5] A. Kimura, *et al.*, "Performance of a high speed and high density data acquisition system for multiple gamma-ray detection," *Conference Record of IEEE NSS/MIC/RTDS 2008*, Article Number: N30-68 (2008).

[6] R. L. Macklin, J. A. Harvey, N. W. Hill, "Neutron Transmission Measurement and Resonance Analysis of ⁹³Zr from 60 to 6000 eV," *Nucl. Sci. Eng.*, **92**, 525 (1986).