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Neutron Capture Cross Section Measurement on ^{91}Zr at J-PARC/MLF/ANNRI

Jun-ichi Hori[1,a], Hiroshi Yashima[1], Shoji Nakamura[2], Kazuyoshi Furutaka[2], Kaoru Y. Hara[3], Hideo Harada[2], Kentaro Hirose[2], Masayuki Igashira[3], Tatsuya Katabuchi[3], Atsushi Kimura[2], Koichi Kino[4], Fumito Kitatani[2], Yoshiaki Kiyanagi[5], Mitsuo Koizumi[2], Motoharu Mizumoto[3], Tadafumi Sano[1], Yoshiyuki Takahashi[1], and Yosuke Toh[2]

[1]*Kyoto University, Research Reactor Institute, 590-0494 Kumatori-cho, Osaka, Japan*
[2]*Japan Atomic Energy Agency, Nuclear Science and Engineering Center, 319-1195 Tokai-mura, Ibaraki, Japan*
[3]*Tokyo Institute of Technology, Research Laboratory for Nuclear Reactors, 152-8550 O-okayama, Tokyo, Japan*
[4]*Hokkaido University, Faculty of Engineering, 060-8628 Sapporo, Hokkaido, Japan*
[5] *Nagoya University, Graduate school of Engineering, 464-8603 Furo-cho, Chikusa-ku, Nagoya, Japan*

Abstract. The neutron capture cross section measurement on ^{91}Zr was performed at neutron TOF beam line ANNRI installed at J-PARC/MLF. Prompt capture gamma rays from the sample were detected with an array of large Ge detectors at a distance of 21.5 m from the spallation neutron source by the time-of-fligh (TOF) method. The neutron capture gamma-ray pulse-height spectra from the 182-eV p-wave resonance and the 292-eV s-wave resonance were obtained by gating on the TOF regions, respectively. Though the decay patterns of primary transitions from the capture state were quite different between resonances, the prominent characteristics common to both resonances was the very strong ground-state transition from the 935-keV state. Therefore, a ground-state transition method was applied to obtain the capture yield, so that the background components due to impurities were successfully eliminated. The preliminary result of the neutron capture cross section for ^{91}Zr up to 5 keV is presented.

1 Introduction

Accurate neutron capture cross sections of long-lived fission products (LLFPs) are required for the nuclear transmutation study. Zirconium-93 with a long half-life of 1.53×10^6 year and large cumulative fission yield is one of the most important LLFPs [1]. The neutron capture cross sections of stable zirconium isotopes are also important because those produced in fission reactors accompany ^{93}Zr to the transmutation system and affects the transmutation performance. Furthermore, zirconium alloy which has very low neutron capture cross sections in thermal energy region, high hardness and corrosion resistance is largely used as a cladding material of fuel rods in nuclear reactors. The neutron capture cross sections of stable zirconium isotopes also contribute the neutron balance in the reactor.

From the viewpoints mentioned above, a systematic measurement of neutron capture cross sections and capture gamma-ray spectra of zirconium stable isotopes as well as ^{93}Zr has been started. This paper presents the preliminary results of the measurement on ^{91}Zr. Since ^{91}Zr is around the closed shell of ^{90}Zr with a double-magic number, the neutron capture gamma-ray transition with single-particle behaviour can be expected.

2 Experiments

The measurements were performed using Accurate Neutron-Nucleus Reaction measurement Instrument (ANNRI) installed at the neutron TOF beam line No.4 (BL04) of the Material and Life Science Experimental Facility (MLF) in the Japan Proton Accelerator Research Complex (J-PARC) [2, 3]. A large Ge-detector array called as "4πGe spectrometer" [4, 5], which consists of two cluster-Ge detectors, eight coaxial Ge detectors and BGO Compton-suppression detectors, was used for measurement of neutron capture gamma rays from the sample. The 4πGe spectrometer is located at a distance of 21.5 m from the neutron source in the MLF. In this work, we used only two cluster-Ge detectors and BGO Compton-suppression detectors. Each sample was set at the center of the spectrometer. Signals from each Ge crystal were analyzed with digital processors and recorded event by event. In order to make a dead time correction, random timing pulses were fed into each pre-amplifier [6].

The pulsed neutron beam was produced from the spallation reactions in a mercury target irradiated by 3-GeV protons at a repetition rate of 25 Hz. The proton beam power was about 290 kW. The neutron beam was collimated to a diameter of 15 mm at the sample position using a rotary collimator system [7]. The pulsed neutron

[a] Corresponding author: hori@rri.kyoto-u.ac.jp

beam was supplied in the double-bunched mode. The pulse width was 100 ns and the interval between double-bunched pulses was 600 ns.

A ^{91}Zr enriched oxide powder was pressed and sintered to a disk shape with a diameter of 5 mm. The weight of the sample was 78.9 mg. The isotopic purity was 88.7 % as listed in Table 1. Chemical impurities of tungsten and hafnium with 90 and 50 ppm were contained in the sample, respectively. A NaCl sample in diameter of 5 mm and with a weight of 500 mg was also used for energy calibration and determination of the full-energy-peak efficiency of the spectrometer. Each sample was sealed in a bag of fluorinated ethylene propylene (FEP) film. The total measuring time of ^{91}Zr was about 41 hours.

Table 1. Isotopic abundances of the ^{91}Zr enriched sample.

	^{90}Zr	^{91}Zr	^{92}Zr	^{94}Zr	^{96}Zr
Isotopic composition (%)	6.1	88.7	3.4	1.5	0.3

3 Analysis and discussion

TOF spectra of ^{91}Zr with and without dead-time correction are shown in Fig. 1. The neutron time-of-flight channel in a horizontal axis is converted into the neutron energy. In the spectrum of ^{91}Zr, we can observe clearly eight resonances at 182, 240, 292, 449, 681, 893, 1531 and 1954 eV. Unfortunately, the resonances above 300 eV are split into two peaks due to the double-bunched-beam effect. The resonance peaks observed below 100 eV are caused by the chemical impurities of tungsten and hafnium. Dips are observed in the low energy side of large resonances. This is because a high count-rate around the peak makes dead time increasing. The dips were removed by dead-time correction method as described in Ref. [6]. The dead time becomes very large above 5 keV as shown in Fig. 1. Therefore, the upper limit of the measurement of neutron capture cross section was set to 5 keV in this work.

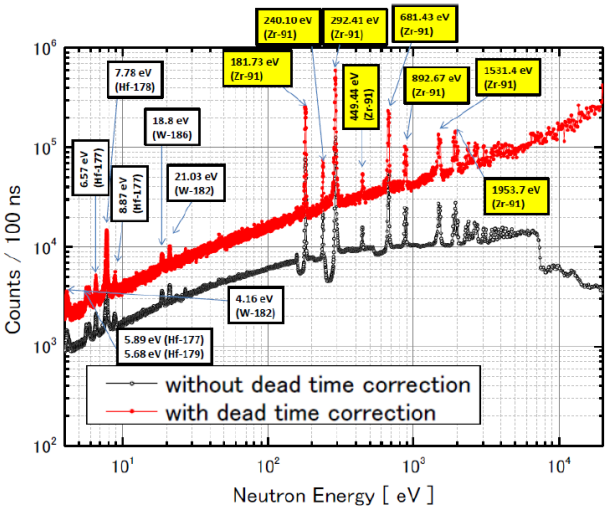

Figure 1. TOF spectra with and without dead-time correction for ^{91}Zr sample.

Figure 2 shows TOF spectra with and without dead-time correction around 292- and 182-eV resonances. Two gates were set in the resonance regions of the TOF spectrum to obtain the pulse-height spectra for the 292- and 182-eV resonances. Another gate was set to the off-resonance region. The net gamma-ray pulse-height spectra corresponding to the 292- and 182-eV resonances were obtained as shown in Figs. 3 and 4.

The prominent characteristics common to both resonances was the very strong ground-state transition from the 935-keV state as shown in Fig. 3. It indicates that almost gamma-ray transitions occur via the 935-keV state. Other cascade transitions to the 935-keV state were also observed in low energy region. Primary transitions from the capture state were observed above 4 MeV. The quite differences of the decay pattern were found between the resonances. We gated on the 935-keV gamma-ray peak and derived the net TOF spectrum for ^{91}Zr capture events as shown in Fig. 5. The resonance peaks due to impurities were almost eliminated and the net TOF spectrum was obtained with a good signal-to-noise ratio.

A neutron spectrum at the sample position was deduced from the previous work performed by detecting the 478-keV gamma rays emitted from the ^{10}B(n,αγ)^{7}Li reaction [8]. The obtained neutron capture cross section of ^{91}Zr is shown in Fig. 6. The evaluated values of JENDL-4.0 [9] are shown together for comparison.

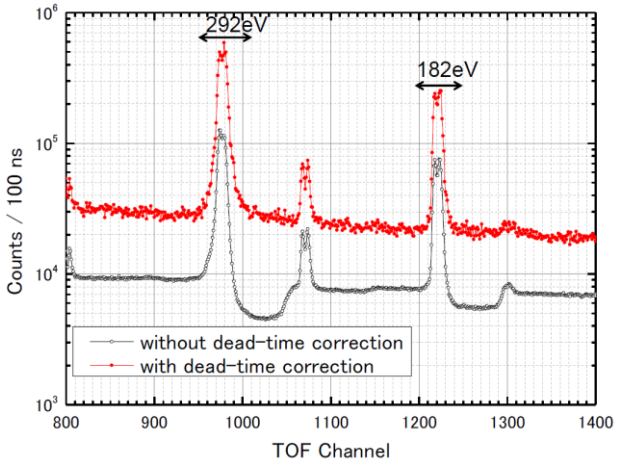

Figure 2. TOF spectra with and without dead-time correction for ^{91}Zr sample.

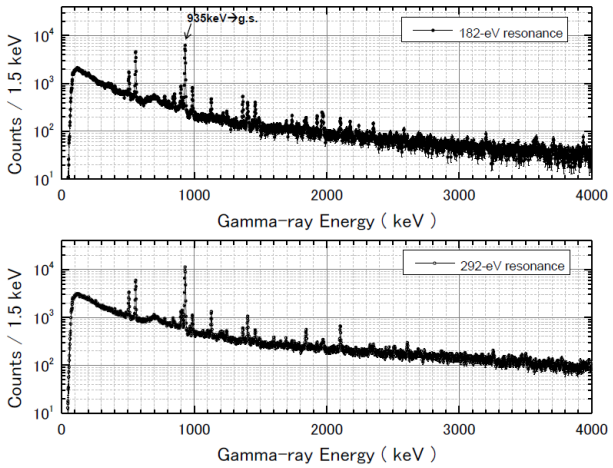

Figure 3. Low energy part of net gamma-ray pulse-height soectra for the 182- and 292-eV resonances.

For comparison in the resonance energy region, the doublet peaks originating from the double-bunched beam operation of J-PARC should be considered. Resonance analysis to internalize the beam structure is in progress. However, the present work indicates that the ground-state transition method is feasible for neutron capture cross section measurement of ^{91}Zr.

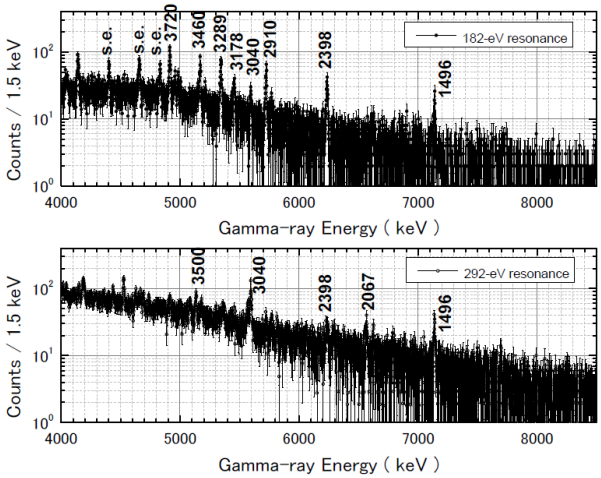

Figure 4. High energy part of net gamma-ray pulse-height soectra for the 182- and 292-eV resonances. Single-escape peaks are indicated with "s.e.". Primary transitions are indicated with the energies of final states.

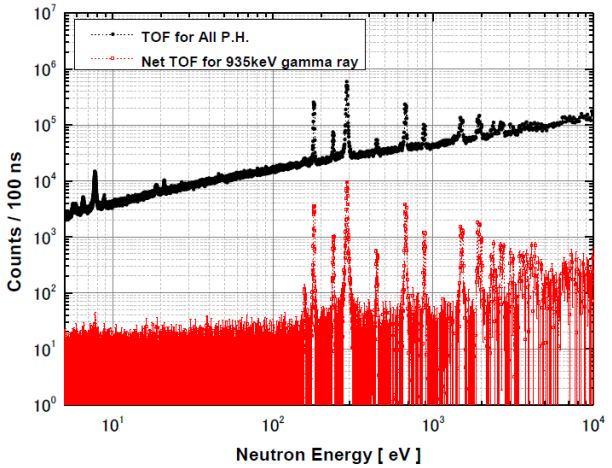

Figure 5. Comparison with the TOF spectra for all range (black line) and the net area of the 935-keV peak (red line) in the pulse-height spectrum of ^{91}Zr.

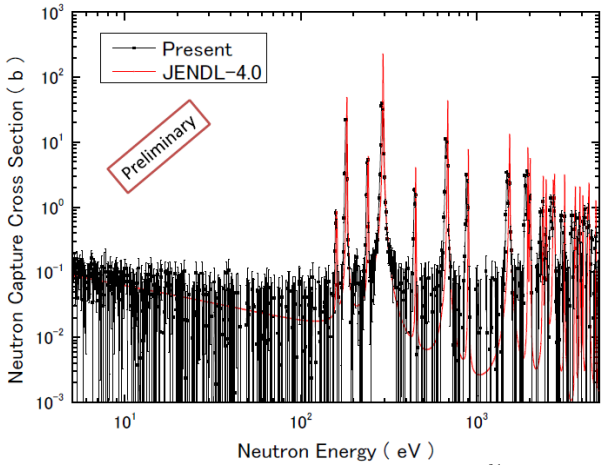

Figure 6. Neutron capture cross section of ^{91}Zr.

4 Summary

The neutron capture gamma-ray pulse-height spectra from the 182- and 292-eV resonances were derived for the first time. The very strong ground-state transition from 935-keV state was observed. By using the ground-state transition, the preliminary results of the neutron capture cross section of ^{91}Zr was obtained up to 5 keV. The background due to impurities contained in the sample was successfully eliminated and the signal-to-noise ratio of neutron capture yield was improved.

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