Neutron Capture Cross Section Measurement and Resonance Analysis of ^{107}Pd Using ANNRI at MLF/J-PARC

Hideto Nakano[1,*], *Tatsuya* Katabuchi[1], *Kazushi* Terada[2], *Atsushi* Kimura[3], *Shoji* Nakamura[3], *Shunsuke* Endo[3], *Gerard* Rovira[3], and *Yu* Kodama[3]

[1]Laboratory for Zero-Carbon Energy, Institute of Innovative Research, Tokyo Institute of Technology, Japan
[2]Institute for Integrated Radiation and Nuclear Science, Kyoto University, Japan
[3]Nuclear Science and Engineering Center, Japan Atomic Energy Agency, Japan

Abstract. The neutron capture cross sections of ^{107}Pd were measured using the Accurate Neutron-Nucleus Reaction Measurement Instrument (ANNRI) at the Japan Proton Accelerator Research Complex (J-PARC). Capture γ-rays were detected with a NaI(Tl) spectrometer. The incident neutron energy was determined by the time-of-flight method. The preliminary cross section was derived from thermal to resonance energy region. In the resonance region, resonance parameters were derived by resonance analysis using REFIT code. The resonance parameter was close to the value of JENDL-4.0.

1 Introduction

The long-term accumulation of Long-Lived Fission Products (LLFPs) in nuclear waste has been a significant issue in nuclear industry due to their long half-lives. The nuclear transmutation of LLFPs into short-lived or stable nuclides is is an attractive option to reduce the current amount of high-level radioactive waste[1]. LLFPs nuclear transmutation systems need highly accurate nuclear data for the neutron-induced nuclear reactions. However, the accuracy of neutron capture cross sections of LLFPs does not meet the requirement for designing the transmutation system[2]. Palladium-107 (half life: 6.5×10^6 y) is one of the important LLFPs, and accurate data for the neutron capture cross section are needed for the study on LLFPs transmutation systems. Nevertheless, only a few experiments to measure the neutron capture cross section of ^{107}Pd have been performed. The neutron energy regions of most of the measurements are limited.

Palladium-107 has the half-life of 6.5 million years and is a pure beta-decay nuclide. The neutron capture reaction of ^{107}Pd produces the stable isotope ^{108}Pd. Thus, the activation method cannot be applied for cross section measurements. Figure 1 shows previously reported experimental data[3][4] and cross sections of evaluated nuclear data libraries. Resonance parameters were measured about 40 years ago and the resonance parameters in evaluated nuclear data libraries are based on these experimental results. The thermal neutron capture cross section was given by only single experiment conducted by Nakamura[5]. JENDL-4.0[6] adopted the experimental value of the thermal cross section in the evaluation while ENDF/B-VIII[7] did not. This causes a large difference of the cross section between the two nuclear data libraries at the thermal neutron energy. To reduce the uncertainties of the neutron capture cross section of ^{107}Pd, new measurements for a wide energy range from the thermal to keV energies are needed. We have previously conducted measurements of the neutron capture cross section of ^{107}Pd at the Japan Proton Accelerator Research Complex (J-PARC) [8]. However, the J-PARC accelerator was in the early stage and the beam power was low, resulting in low count statistics. In addition, background measurements were incomplete due to the limited beam time of the previous measurement. The beam power of J-PARC was increased six folds in the past decade and the current beam power reaches 700 kW. Using the high intensity neutron beam, a new measurement was conducted. The results of the new measurement will be given in the present report.

2 Experimental setup

The neutron capture cross section measurements were carried out using the Accurate Neutron Nucleus Reaction Measurement Instrument (ANNRI)[9] at the Materials and Life Science Facility (MLF) of J-PARC. A high intensity pulsed neutron beam from the Japanese Spallation Neutron Source at MLF was used. Caputure γ-rays were detected with a NaI(Tl) detectors of ANNRI. The time-of-flight (TOF) method was employed to determine the incident neutron energy. The TOF and the pulse-height (PH) of each event were acquired and recorded. The crystal size of the NaI(Tl) spectrometer was 330 mm in diameter and 203 mm in thickness. The detector was located at an angle of 90 degrees with respect to the beam axis. The neutron flight length was 27.924 m. The J-PARC accelerator was operated in the double bunch mode at a beam power of

*e-mail: nakano.h.ag@m.titech.ac.jp

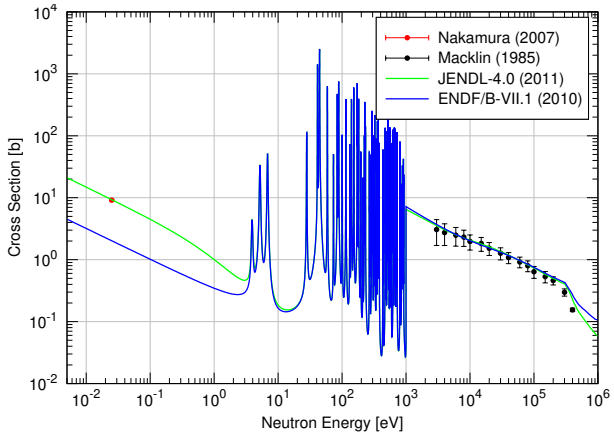

Figure 1. Current status of neutron capture cross section dataset and evaluated nuclear data of ^{107}Pd.

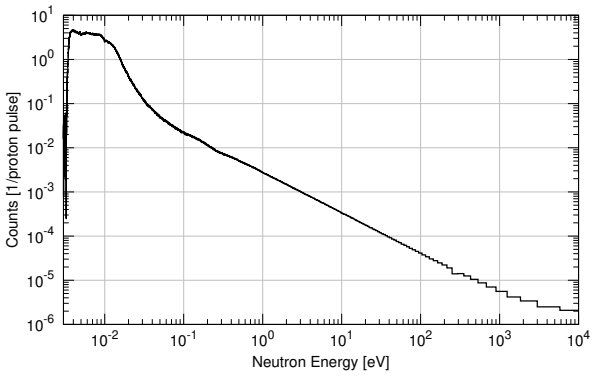

Figure 2. Neutron spectrum derived from 478-keV γ-ray counts of the ^{10}B(n,$\alpha\gamma$)^{7}Li reaction..

730 kW. The total mass of ^{107}Pd sample was 137.4 mg and the isotopic ratio of ^{107}Pd was 15%. Thus, the net weight of ^{107}Pd was 21.0 mg. The sample was capsulated in an aluminum container. In addition to the sample measurement runs, a dummy case which was identical to the sample aluminum container was measured for background measurement. Measurements without sample were also made for blank background measurement. A carbon sample was measured to estimate sample-scattering neutron background. Samples of the stable isotopes, ^{105}Pd, ^{106}Pd, ^{108}Pd were measured to derive contributions from isotope impurities. The stable isotope abundances of ^{105}Pd, ^{106}Pd, ^{108}Pd in the ^{107}Pd sample were 48wt%, 23wt% and 9wt%, respectively. A B_4C sample was measured to obtain the neutron energy spectrum from 478 keV gamma-ray counts of the ^{10}B(n,$\alpha\gamma$)^{7}Li reaction. Measurement of a gold sample was made as standard measurement.

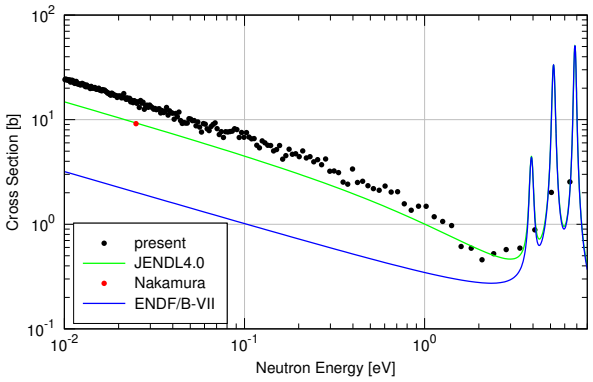

Figure 3. The preliminary capture cross section below resonance region.

3 Results

The neutron spectrum was derived from detected counts of 478-keV γ-rays from the ^{10}B(n,$\alpha\gamma$)^{7}Li reaction in the B_4C run. Corrections for the self-shielding and multiple scattering effects in the sample were estimated with the Monte-Carlo simulation code PHITS[10]. The neutron spectrum in this work is shown in Figure 2.

The capture cross section below the resonance region was derived. Dead time collection was applied to each spectrum and frame overlap background was removed. Blank, neutron scattering, aluminum case background were subtracted. Finally, the stable isotope impurity contributions were removed. The obtained net ^{107}Pd yield was divided by the number of the incident neutrons derived from the B_4C measurement. The relative cross section was normalized to the evaluated value of JENDL-4.0 at the 44-eV resonance of ^{107}Pd. The obtained value is shown in Figure 3. The thermal cross section value in this work was 1.6 times higher than that of JENDL-4.0. The evaluated value is based on the experimental value of Ref.[5] that was derived by the ground-state transition method. The ground-state transition method gives only a lower limit of the capture cross section. The disagreement between the present value and the JENDL-4.0 evaluation may come from possible transition missing in the ground-state transition method.

Resonance analysis was conducted with REFIT code[11] in the resonance region. A part of fitted result is shown in Figure 4. Forty resonances were observed in the energy region from 1 eV to 500 eV and 16 resonances were fitted by REFIT. The average Γ_γ in this work was 0.154 eV, which was close to JENDL-4.0.

4 Conclusion

Highly accurate nuclear data of ^{107}Pd for the neutron-induced nuclear reactions are required for the design of LLFPs nuclear transmutation systems. The neutron capture cross section measurement of ^{107}Pd was conducted using the Accurate Neutron-Nucleus Reaction Measurement Instrument at the Japan Proton Accelerator Research Complex. The incident neutron energy was measured by

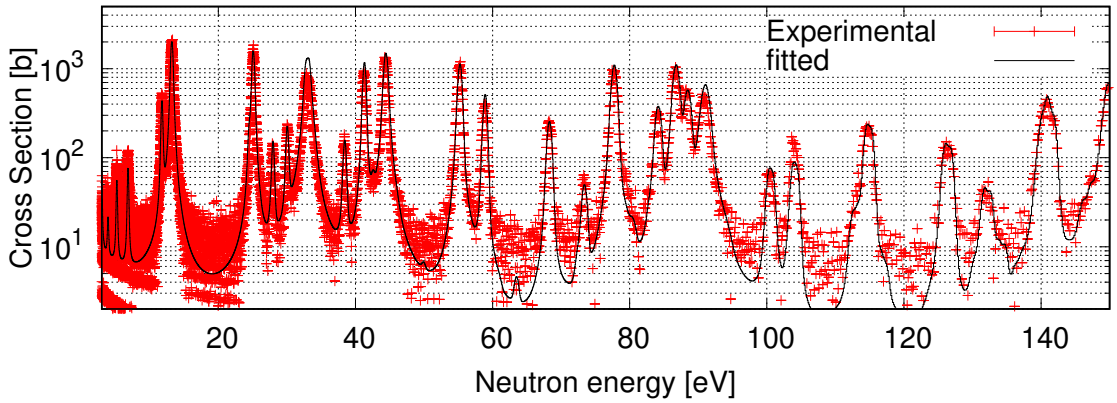

Figure 4. A part of the fitting results by resonance analysis using REFIT code.

the time-of-flight method. The preliminary cross sections were given below 500 eV. In the thermal energy region, the present cross section was about 1.6 times higher than the previous data. In the resonance region, resonance parameters were determined by resonance analysis using REFIT code. The obtained values of the resonance parameters were close to those of JENDL-4.0.

References

[1] T. Wakabayashi et al., Scientific Report **19156**, (2019).

[2] N. Yamano et al., Journal of Nuclear Science and Technology **58** 567-578, (2020).

[3] R. L. Macklin, Nuclear Science and Engineering **89** 79-86, (1985).

[4] U. N. Singh et al., Nuclear Science and Engineering **67** 54-60, (1978).

[5] S. Nakamura et al., Journal of Nuclear Science and Technology **44** 103-108, (2007).

[6] K. SHIBATA et al., Journal of Nuclear Science and Technology **48** 1-30, (211).

[7] M.B.Chadwick et al., Nuclear Data Sheets **107** 2931-3060, (2006).

[8] K. Terada et al., Progress in Nuclear Energy **82** 118-121, (2015).

[9] K. Kino et al., Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment **626-627** 58-66, (2011).

[10] T. Sato et al., Journal of Nuclear Science and Technology **55** 684-690, (2017).

[11] M. C. Moxon et al., Users ' Guide for REFIT-2009-10 UKNSF 243, (2010).