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ARTICLE

Neutron capture cross section measurement of Palladium-107 using J-PARC

Hideto Nakano [a]*, Tatsuya Katabuchi [a], Kazushi Terada [c], Atsushi Kimura [b], Shoji Nakamura [b], Shunsuke Endo [b], Gerard Rovira [b] and Yu Kodama [a]

[a] *Laboratory for Zero Carbon Energy, Tokyo Institute of Technology;* [b] *Nuclear Science and Engineering Center, Japan Atomic Energy Agency;* [c] *Institute for Integrated Radiation and Nuclear Science, Kyoto University*

Neutron capture cross section measurements for ^{107}Pd using an intense neutron beam from a spallation neutron source with a wide energy range from thermal to keV regions have been conducted. A neutron detector was placed upstream of the beam line to monitor the number of the incident neutrons. A time-of-flight (TOF) method using a NaI(Tl) detector was employed and the data were analyzed based on the pulse-height weighting technique in order to derive the neutron capture cross-section. An energy-dependence of cross-section was obtained relative to the incident neutron spectrum derived from the ^{10}B(n,$\alpha\gamma$)^{7}Li reaction yield. The present cross section is about two times larger than the past experimental value in the thermal energy region. The present results and the past experimental values are in good agreement in the keV energy region. In the resonance region, resonance analysis was performed using REFIT code. The average value of radiation width of s-wave resonances was 0.181 ± 0.009 eV.

Keywords: ***ong-lived fission products (LLFPs); Palladium-107; neutron capture cross section; Japan Proton Accelerator Research Complex (J-PARC); time-of-flight method***

1. Introduction

The nuclear industry has an issue on disposal of long-lived fission products (LLFPs) in nuclear waste that have been accumulated due to their long half-lives. The nuclear transmutation of LLFPs into short-lived or stable nuclides is expected as a solution to reduce the current amount of high-level radioactive waste. Highly accurate nuclear data for the neutron-induced nuclear reactions are necessary in order to design LLFPs nuclear transmutation systems.

Study of transmutation of minor actinides (MAs) have been conducted by different research groups. On the other hand, transmutation of LLFPs has not been studied intensively compared to MAs because the priority of transmutation of LLFPs has been thought be lower. However, in addition to the design of a nuclear transmutation system for LLFPs themselves, neutron reaction data of LLFPs are important in transmutation systems for MAs. In the transmutation systems, MAs are transmuted via the neutron-induced fission reaction, through which LLFPs are generated and accumulated. Generated LLFPs are transmuted through the neutron capture reaction. Balance between the production and the consumption of LLFPs in the core determines the performance of a nuclear transmutation system. Yamano et al. estimated the uncertainties of the transmutation rates of LLFPs in a fast nuclear reactor [1]. The estimated uncertainties of transmutation rates are large, especially for ^{79}Se, ^{93}Zr, ^{107}Pd. The large uncertainties of the calculated transmutation rates come from ambiguities of nuclear data of these LLFPs that originate from a smaller number of experimental cross section data. In the present research, therefore, we focus on reducing the uncertainties of the neutron capture cross section of ^{107}Pd.

Current status of the neutron capture cross section of ^{107}Pd is shown in the **Figure 1**. Past experimental data and evaluated cross sections from evaluated nuclear data libraries JENDL-5 [2] and ENDF/B-VIII.0 [3]. At the thermal neutron energy (25.3 meV), only a single measurement by Nakamura et al. [4] exists. In the resonance region, there are two data sets of resonance parameters measured by Macklin [5] and Singh et al. [6]. In the keV region, only one data set of the capture cross section was provided by Macklin [5]. The evaluated cross sections of JENDL-5 and ENDF/B-VIII.0 disagree with each other below 4 eV while they agree in the higher energy region. The JENDL-5 evaluation adopted the thermal cross section value of Ref. [4] but the ENDF/B-VIII.0 evaluation did not. This difference of the evaluations causes the discrepancy of the two evaluated cross sections in the low energy region. In the higher energy region, the two evaluations agree but this is just because they are based on the same experimental data set by Ref. [5] that is almost only available single data set. In order to carry out reliable evaluation of the neutron capture

*Corresponding author. E-mail: nakano.hideto@jaea.go.jp

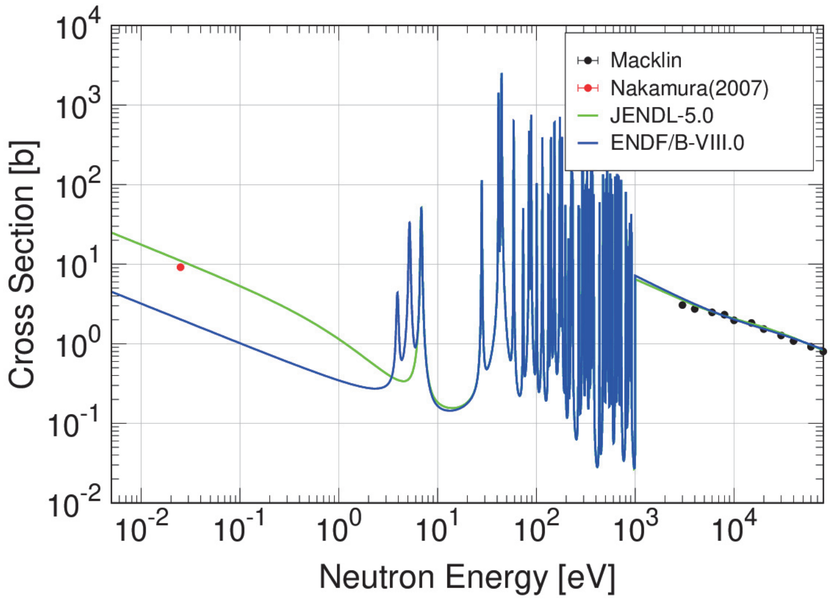

Figure 1. Past experimental data and evaluated values of the neutron capture cross section of ^{107}Pd.

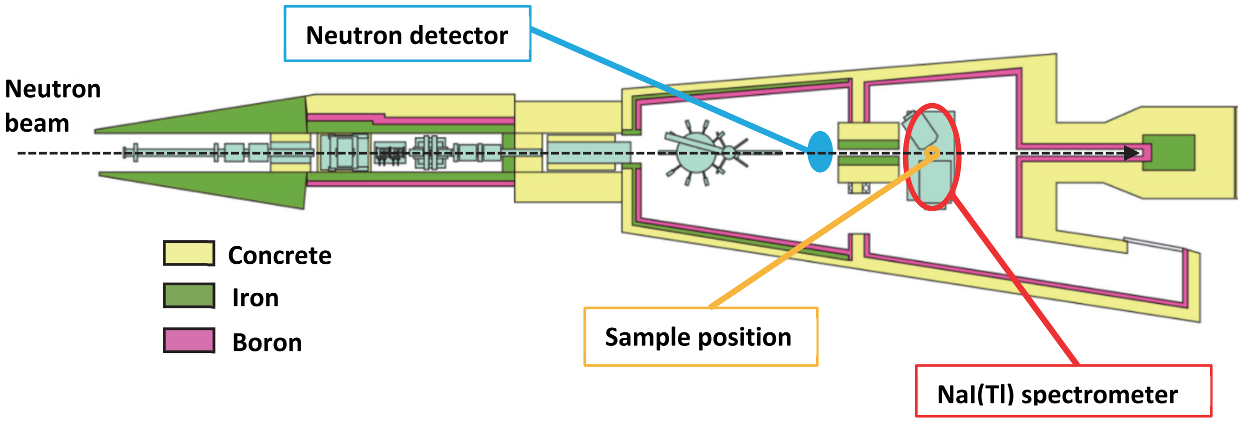

Figure 2. Experimental setup of ANNRI in J-PARC/MLF.

cross section of ^{107}Pd, new independent experiments are required. Thus, we conducted new measurement of the capture cross section for ^{107}Pd using an intense neutron beam from a spallation neutron source with a wide energy range from thermal to keV regions.

2. Experiments

Experiment was performed using a pulsed neutron beam from a spallation neutron source of the Materials and Life Science Facility (MLF) in the Japan Proton Accelerator Research Complex (J-PARC). The Accurate Neutron-Nucleus Reaction Measurement Instrument (ANNRI), a neutron beam line designed for nuclear data measurement, was used. The J-PARC accelerator was operated in the double bunch mode at a beam power of 730 kW and a repetition rate of 25 Hz. **Figure 2** shows experimental setup. The sample was placed at a flight length of 27.9 m from the neutron source. The incident neutron energy was determined by the neutron TOF method. A NaI(Tl) detector at a scattering angle of 90° was used to detect prompt γ-rays from the capture cross section. The NaI(Tl) detector was shielded with a combination of different shielding materials from background γ-rays and neutrons. A neutron detector was placed upstream of the beam line to monitor the number of the incident neutrons. The incident neutron detector was developed specially to withstand high intensity neutron beam from the spallation source of J-PARC [7]. The detector consisted of a thin ^{6}Li foil and a thin plastic scintillator. Neutron detection material was an enriched ^{6}LiF foil. Neutrons react with ^{6}Li, causing the ^{6}Li(n,t)^{4}He reaction. Triton and alpha particles enter scintillator, converting their deposited energies to scintillation photons which are detected with a photo multiplier tube. The detected neutron counts were used for normalization of measurements.

The Pd sample was a metal pellet cased in an aluminum container as shown in **Figure 3**. The net weight of ^{107}Pd was 21.0 mg and the radioactivity was 398 kBq. The sample contained other palladium isotopes. The isotope ratios of palladium isotopes in the sample are shown in **Table 1**. While the abundance ratio of ^{107}Pd was only 15%, the ^{105}Pd abundance is 48%. This means that the background evaluation of isotope impurities is significantly important. In addition to the Pd sample, isotope enriched samples of ^{105}Pd, ^{106}Pd and ^{108}Pd were measured to estimate the impurity background components.

To evaluate other background components, other measurements were made. A dummy case sample was measured for evaluation of the aluminum case background, a carbon sample for scattering neutron background, blank measurement for sample-independent background.

The incident neutron energy spectrum was derived from counts of 478 keV γ-rays from the ^{10}B(n,αγ)^{7}Li reaction when a boron carbide sample was placed at the sample position. The absolute value of the capture cross section was determined by the saturated resonance method using a gold sample.

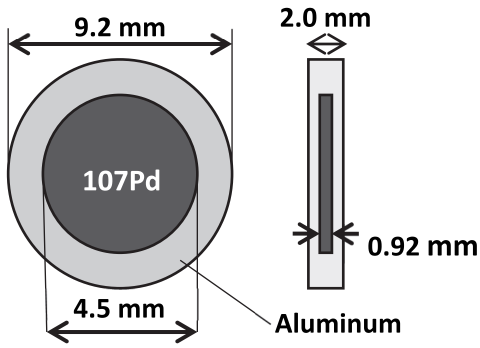

Figure 3. Pd sample.

Table 1. Isotope abundance of the ^{107}Pd sample.

Isotope	104	105	106	107	108	110
Ratio(%)	2	48	23	15	9	3

3. Results and discussion

Neutron capture yield of ^{107}Pd was calculated using the pulse height weighting technique. Neutron binding energy of ^{107}Pd is 9.2 MeV, hence gamma-ray events with energies lower than 9.2 MeV were used to derive the neutron capture yield. Several backgrounds were estimated and removed from foreground spectrum. In particular, evaluation of the Pd isotope background is significant as mentioned in the sample description. The effects of ^{105}Pd, ^{106}Pd and ^{108}Pd were estimated by measuring isotope enriched samples. Weight discrepancy between the Pd sample and enriched samples were corrected with ratio of sample weight. After background removal and correction of neutron multiple scattering and self-shielding, net capture yield of ^{107}Pd was divided by the number of the incident neutrons derived from counts of 478 keV γ rays from ^{10}B(n,αγ)^{7}Li reaction. Finally, to derive the absolute cross section, the relative ratio of the capture yield to the neutron count was normalized to the saturated resonance of ^{197}Au at 4.9 eV.

The obtained neutron capture cross section of ^{107}Pd in the thermal energy region is shown in **Figure 4**. The present result is compared with the experimental value of Ref. [4] and evaluated cross sections of JENDL-5 and ENDF/B-VIII.0. The cross section of the present work is about two times larger than the past experimental value. A method of experiment and analysis in previous study were different from ours. However, it was difficult to obtain the more detail information and compare the data. Therefore, the origin of the discrepancy is not clear. Further investigation is needed. **Figure 5** shows the present results in the keV region. Experimental values of Ref. [5] and evaluated values of JENDL-5 and ENDF/B-VIII.0 are also plotted. The present results and the past experimental values are in good agreement.

In the resonance region, resonance analysis was performed using REFIT code [8] for the analysed experimental data. Resonance parameters taken from JENDL-5 were inputted as initial values. In the neutron energy range from 1 to 500 eV, 17 resonances were analysed to derive the resonance parameters. Examples of the fitting results are shown in **Figures 6-8**. The red points represent the experimental data and the black line is the result of the fitting process by REFIT. The average value of radiation width of s-wave resonances was 0.181 ± 0.009 eV.

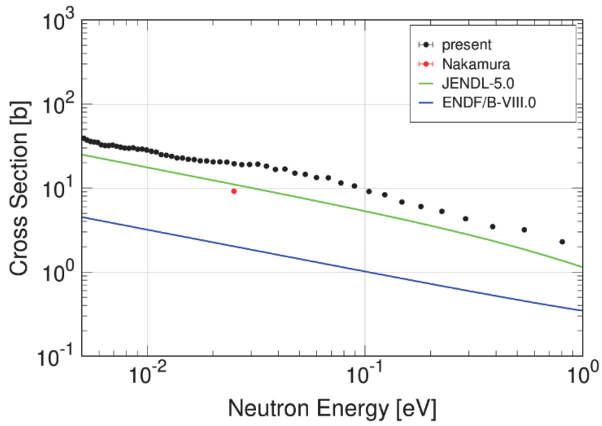

Figure 4. Neutron capture cross section of ^{107}Pd in the thermal energy region.

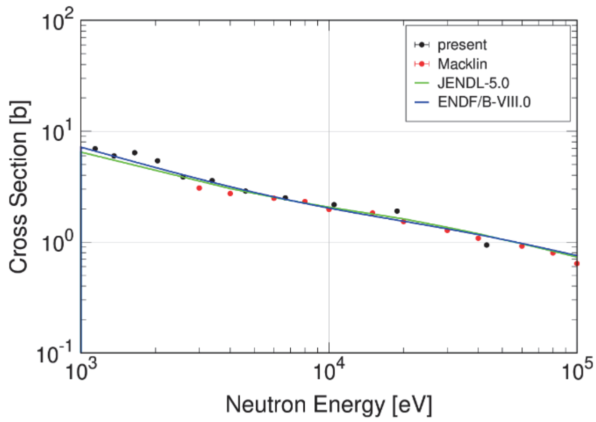

Figure 5. neutron capture cross section of ^{107}Pd in the keV region.

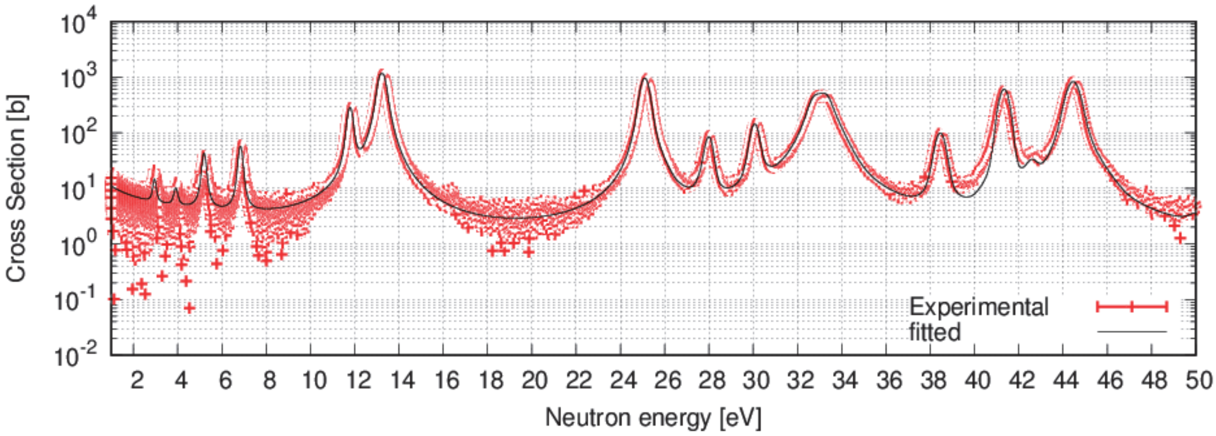

Figure 6. Fitting results in the resonance analysis from 1 to 50 eV.

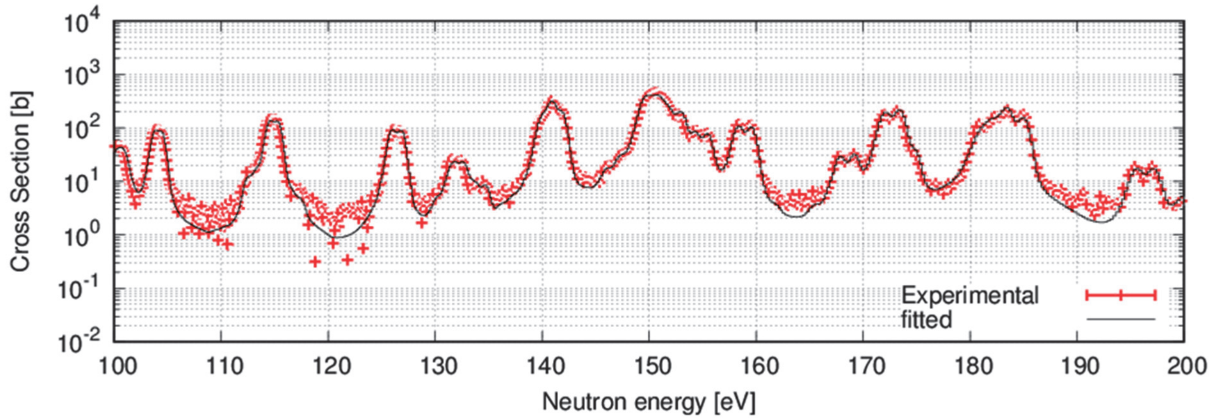

Figure 7. Fitting results in the resonance analysis from 100 to 200 eV.

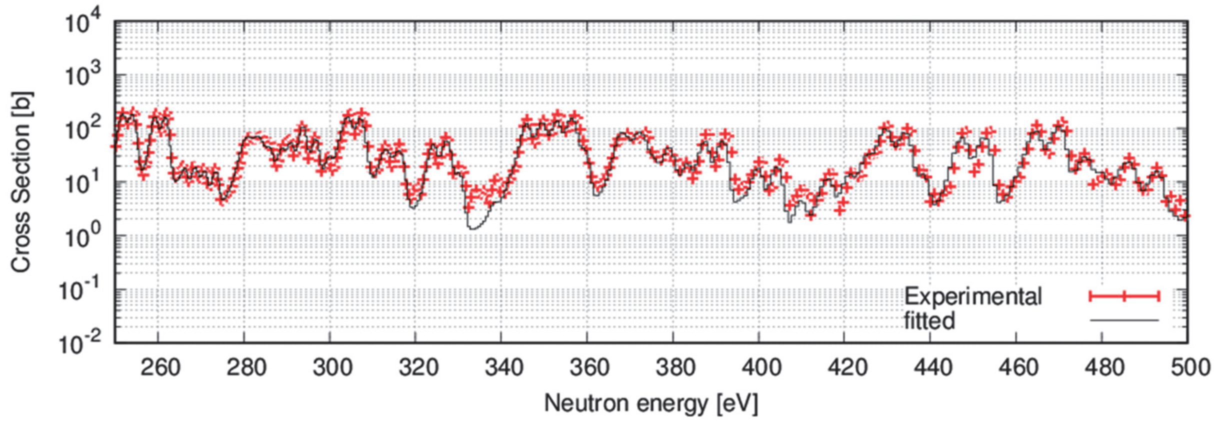

Figure 8. Fitting results in the resonance analysis from 250 to 500 eV.

4. Conclusion

A new measurement of the neutron capture cross section for ^{107}Pd was carried out in a wide energy range from the thermal to keV energy regions at ANNRI of J-PARC/MLF. The present cross sections in the thermal energy region were about two times higher than the past experimental value. In the keV energy region, the present result and the past measurement agree well. Resonance analysis was carried out in the neutron energy range from 1 to 500 eV. The resonance parameters of 17 resonances were determined.

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