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Measurement of neutron capture cross sections of Pd-107 at J-PARC/MLF/ANNRI

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A B S T R A C T

Measurements of the neutron capture cross sections of ^{107}Pd were carried out at the Materials and Life Science Experimental Facility (MLF) of the Japan Proton Accelerator Research Complex (J-PARC). Gamma-rays were detected with an NaI(Tl) spectrometer of the Accurate Neutron–Nucleus Reaction Measurement Instrument (ANNRI). The neutron capture cross sections were determined by the time-of-flight method in the neutron energy region from the thermal to keV energies.

1. Introduction

Long-lived fission products (LLFPs) produced in nuclear fission reactors are problematic for future nuclear waste management. Nuclear transmutation of LLFPs into stable or short-lived nuclides by neutron capture reaction is an attractive option to reduce the environmental burden in the geological disposal of nuclear waste (Takano and Ikegami, 2002).

Palladium-107 (half-life: 6.5×10^6 y, fission yield for U-235: 1% and Pu-239: 3%) is one of the most important LLFPs. Its neutron capture cross section is necessary for the study of LLFP transmutation systems. However, there are only a few previous measurements for the capture cross section of ^{107}Pd. Singh et al. carried out the neutron capture and transmission measurements using an electron-accelerator neutron source of the Rensselaer Polytechnic Institute Gaerttner Laboratory (Singh et al., 1978). The resonance parameters of 34 resonances below 700 eV were determined. Macklin made neutron capture measurements at the Oak Ridge Electron Linear Accelerator facility (Macklin, 1984). He determined the resonance parameters of 130 resonances up to 3.5 keV and the average capture cross sections in the energy region from 3 to 600 keV. Nakamura et al. performed thermal neutron capture measurement at the 8-MW Los Alamos Omega West Reactor with a prompt γ ray spectroscopic method, and determined a lower limit of the thermal neutron capture cross section (Nakamura et al., 2007). The evaluated cross sections in JENDL-4.0 and ENDF/B-VII.1 above 3 eV are close together because the evaluated data are based on mainly the Macklin's results. However a large difference exists between the evaluations in the lower neutron enegies. It comes from the difference of the adopted thermal capture cross section values. Recently, in order to improve the reliability of the cross section data,the capture cross section of ^{107}Pd was measured with a large Ge detector array in the neutron energies from the thermal to a few hundred eV by the time-of-flight (TOF) method at the Japan Proton Accelerator Research Complex (J-PARC) (Nakamura et al., to be published). In the present work, we

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measured the capture cross section of ^{107}Pd with a faster NaI(Tl) detector system installed in the J-PARC beam line to extend the high energy limit of measurement.

2. Experiments

Experiments were carried out at the Materials and Life Scei-nece Experimental Facility (MLF) of J-PARC. Neutrons were produced via the spallation reaction induced by a 3-GeV proton beam impinging on a mercury target of MLF. The accelerator was operated at a repetition rate of 25 Hz. The proton beam power on the spallation target was 120 kW. The produced neutrons were moderated in liquid hydrogen moderators cooled at 19 K, thereby providing neutron beams having wide energy neutron spectrum (Maekawa et al., 2010). Signal from a proton beam pulse monitor located in a beam transport tube to the spallation target was used as the start trigger for TOF measurement. The number of proton beam pulses was recorded and used for normalization of each run.

The accelerator was run in a special operational mode called “single bunch” mode in the present experiments. In normal operation, two proton bunches of 100 ns pulse width, separated at an interval of 600 ns, are injected into the spallateion target for each neutron burst. This is beneficial to increase the neutron flux in the thermal and cold neutron energy regions where the double bunch time structure of the proton beam disappears after the moderation process in the moderator. However the double bunch proton beam causes doublet neutron resonance peaks above about 100 eV, requiring complicated procedure for resonance analysis. The single bunch operation in the present experiments was approved for our special request to solve this complicated issues in the double bunch mode and obtain basic data for future analysis of experimental data acquired in the double bunch operation.

The characteristics of the samples are summarized in Table 1. ^{107}Pd metal powder was formed into a pellet and then sealed in an aluminum container. The weight of the pellet was 137 mg. The isotopic composition of the ^{107}Pd sample was determined with a Thermal Ionization Mass Spectrometer. The determined isotopic purity of ^{107}Pd was 15.3%. The net weight of ^{107}Pd is 21.0 mg from the isotopic purity. Stable palladium isotopes existed in the ^{107}Pd sample. Thus, in addition to the ^{107}Pd sample run, measurements for enriched samples of ^{105}Pd and ^{108}Pd listed in Table 1 were made to estimate the contribution from the stable isotopes in the ^{107}Pd sample. ^{106}Pd was also one of the major isotopic contaminants but an experimental run for an enriched ^{106}Pd sample was not done because of a limited beam time of the single bunch mode. Instead, the measurement for ^{106}Pd was performed in a different beam time operated in the normal double bunch mode for future analysis.

Table 1
Characteristics of samples.

Sample	^{105}Pd	^{107}Pd	^{108}Pd
Chemical form	Metal	Metal	Metal
Chemical purity [%]	99.97		99.97
Gross weight [mg]	23.4	137.4	9.8
Composition [%]			
^{102}Pd	<0.05	<0.02	<0.02
^{104}Pd	0.45 ± 0.3	1.6 ± 0.4	0.06
^{105}Pd	98.4 ± 0.1	48.4 ± 0.8	0.12
^{106}Pd	1.05	23.1 ± 0.3	0.37
^{107}Pd	–	15.3 ± 0.4	–
^{108}Pd	0.08	8.7 ± 0.3	99.2
^{110}Pd	<0.05	2.6 ± 0.3	0.25
Diameter [mm]	5.0	4.5	5.0
Thickness [mm]	0.02	0.92	0.03

The sample was placed at the sample position of the downstream experimental area of the Accurate Neutron–Nucleus Reaction Measurement Instrument (ANNRI) (Igashira, 2009; Kino et al., 2011). The flight path length from the neutron source was 27.9 m. Neutrons traveled through a carbon fiber beam duct, which housed the sample. Helium gas was regularly run through the beam duct to reduce scattering neutron background from air. Gamma-rays from the sample were detected with an NaI(Tl) spectrometer of ANNRI. The detection angle was 90° with respect to the neutron beam. Signal processing and data acquisition were made based on a pulse width analysis method (Katabuchi et al., to be published). We have developed this method for fast data acquisition using a time digitizer FAST ComTech MCS6A. The TOF and the pulse width of each event were recorded event-by-event in a list-data format file. The pulse width was converted to the pulse height using a conversion curve determined in calibration experiments.

The measuring times were summarized in Table 2. A natural boron sample was used to determine the shape of the incident neutron energy spectrum from the ^{10}B(n, $\alpha\gamma$)^{7}Li reaction. An empty aluminum container (Dummy) identical to the ^{107}Pd sample container was used for background measurement from the Al container. To estimate background of scattering neutrons from the ^{107}Pd sample, ^{208}Pb measurement was made. Blank and ^{208}Pb runs were also made for background estimations for no sample and scattered neutrons, respectively.

3. Data analysis

3.1. Incident neutron energy spectrum

The incident neutron energy spectrum was determined from counts of 478 keV γ-rays from the ^{10}B(n, $\alpha\gamma$)^{7}Li reaction. TOF spectra of gated counts in the 478 keV peak region were made for the boron, ^{208}Pb and blank runs. Sample-independent and sample-scattered neutron backgrounds were estimated from the TOF spectra of ^{208}Pb and blank runs and subtracted from the boron TOF spectrum. The cross section data of the ^{10}B(n, $\alpha\gamma$)^{7}Li reaction in JENDL-4.0 (Chiba, 2010) was used to convert the detected counts to the number of the incident neutrons. The dead time correction was made for the net TOF spectrum. The correction for the neutron self-shielding and multiple scattering in the boron sample was made by calculating the correction factors with the Monte-Carlo simulation code PHITS (Sato et al., 2013). The obtained incident neutron energy spectrum is shown in Fig. 1. The broad peak around 10 meV is the thermalized neutron bump in the liquid hydrogen moderator. The spectrum showed the typical $1/E$ energy dependence from 0.1 eV to 1000 eV. The observed dips above 100 eV are attributed to resonance absorption by the contents in aluminum alloy used for beam windows of the moderator, and the beam duct and other instruments on the beam line. For example, the large dip at 35 keV was caused by the resonance of ^{27}Al.

Table 2
Measuring times.

	Measuring time [h]
^{107}Pd run	13
^{105}Pd run	8
^{108}Pd run	15
Boron run	10
Dummy run	10
^{208}Pb run	10
Blank run	10