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Measurements of the thermal neutron cross-section and resonance integral for the ^{108}Pd(n,γ)^{109}Pd reaction

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ABSTRACT

The thermal neutron capture cross-section (σ_0) and resonance integral (I_0) of the ^{108}Pd(n,γ)^{109}Pd reaction have been measured relative to that of the monitor reaction ^{197}Au(n,γ)^{198}Au. The measurements were carried out using the neutron activation with the cadmium ratio method. Both the samples and monitors were irradiated with and without cadmium cover of 0.5 mm thickness. The induced activities of the reaction products were measured with a well calibrated HPGe γ-ray detector. In order to improve the accuracy of the results, the necessary corrections for the counting losses were made. The thermal neutron capture cross-section and resonance integral of the ^{108}Pd(n,γ)^{109}Pd reaction were determined to be $\sigma_{0,Pd}$ = 8.68 ± 0.41 barn and $I_{0,Pd}$ = 245.6 ± 24.8 barn, respectively. The obtained results are compared with literature values and discussed.

1. Introduction

Natural palladium (^{nat}Pd) consists of six stable isotopes, among them the ^{108}Pd with abundance of 26.46% can be used for the production of the medical isotope ^{109}Pd, which is used for tumor therapy [1,2]. The ^{108}Pd is also one of the stable fission products [3],which is considered as an important neutron absorbers for thermal and fast reactors [4]. The neutrons absorbed by the fission products in nuclear reactors represent a significant portion of the total loss of neutrons. Therefore, accurate knowledge of the thermal neutron capture cross-section and resonance integral of the ^{108}Pd(n,γ)^{109}Pd reaction is very important with regards to the production of the β-emitting medical isotope ^{109}Pd and the safety analysis of nuclear reactors [5].

In this work, we have chosen the ^{108}Pd(n,γ)^{109}Pd reaction for studies because the number of measured cross-section data are still limited and their values are relatively large scattered. So far, we have found nine thermal neutron capture cross-sections [6–14], and two resonance integral data [10,15] for the ^{108}Pd(n,γ)^{109}Pd reaction. The previous measured thermal neutron capture cross-section values varied from 7.2 ± 0.5 barn [7] to 14 ± 2 barn [12], differ by about 94%. The values of two reported resonance integrals are 253 barn [10] and 173 ± 9 barn [15], differ by about 46%. The lack of reference data and large discrepancies between their values make it difficult to produce the best value for applications. Therefore, additional measurements of the thermal neutron capture cross-sections and resonance integrals can allow for better comparison of results between studies and for applications.

In the present work, we have measured the thermal neutron capture cross-section and resonance integral of the ^{108}Pd(n,γ)^{109}Pd reaction relative to that of the ^{197}Au(n,γ)^{198}Au reaction by means of the neutron activation with cadmium ratio method. The aim of the present work is to give additional thermal neutron capture cross-section and resonance integral for the ^{108}Pd(n,γ)^{109}Pd reaction with the highest possible accuracy. In order to improve the accuracy of the experimental results, the efforts were made to reduce the experimental errors caused in sample irradiation, radio activity measurement, and data processing, as well as to perform the necessary corrections for the counting losses.

2. Experimental procedure

The present experimental work was performed at the pulsed neutron facility using the 100-MeV electron linac of the Pohang Accelerator Laboratory (PAL). The pulsed neutron facility consists of an electron linac, a water-cooled tantalum (Ta)-target system and a 12-m long time-of-flight (TOF) path [16–19]. The electron beam impinges upon a water-cooled Ta-target and produce bremsstrahlung, which in turn

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Table 1
Characteristics of Pd, Au, and In foils.

Foils	Size (mm)	Thickness (mm)	Weight (g)	Purity (%)
Pd1	17 × 17	0.05	0.1653	99.9
Pd2 (Cd)	17 × 17	0.05	0.1669	99.9
Au1	17 × 17	0.03	0.1653	99.95
Au2(Cd)	17 × 17	0.03	0.1803	99.95
In1	12 (dia.)	0.05	0.0509	99.95
In2	12 (dia.)	0.05	0.0521	99.95
In3	12 (dia.)	0.05	0.0520	99.95
In4	12 (dia.)	0.05	0.0508	99.95
In5	12 (dia.)	0.05	0.0509	99.95
In6	12 (dia.)	0.05	0.0483	99.95

generate photo-neutrons by the (γ,xn) reactions. The distributions of neutrons with and without water moderator were described in our previous works [20,21]. At the water level of 5 cm above the Ta-target surface, the measured cadmium ratios ($CR = (R/R_{Cd})$), where R and R_{Cd} are reaction rates per atom for bare and Cd-covered isotope irradiation, were 2.70 ± 0.05 and 1.45 ± 0.02 for the ^{197}Au and ^{108}Pd radionuclides, respectively.

The palladium and gold foils with the same size were prepared for the activation. The neutron fluence exposed to each palladium and gold foil was extrapolated based on the measured activities of the indium monitors. The characteristics of the activation foils used in our experiment are given in Table 1. For irradiation, palladium, gold and indium foils were placed on the top of the water moderator as shown in Fig. 1, where Pd2(Cd) and Au2(Cd) denote the activation samples covered with a 0.5-mm thick Cd. All the palladium, gold and indium foils were irradiated simultaneously for three hours. The nuclear data for the ^{108}Pd(n,γ)^{109}Pd, ^{197}Au(n,γ)^{198}Au, and ^{115}In(n,γ)^{116m}In reactions and some main nuclear data used in calculations are given in Table 2 [22,26,27]. The thermal neutron cross-section and resonance integral of the ^{108}Pd(n,γ)^{109}Pd reaction were determined from the induced activities of the reaction products.

The activity measurements were performed using a well calibrated HPGe γ-ray detector (ORTEC-GEM-20180-p) coupled to a PC-based multichannel analyzer, where the relative efficiency was 20%, and the energy resolution was 1.8 keV full width at half maximum (FWHM) at the 1332.5-keV γ-peak of ^{60}Co. The efficiency of the HPGe γ-ray detector was determined experimentally using a set of standard gamma sources. The detector efficiency calibration was described elsewhere [23]. In order to avoid the coincidence summing effects, the activity measurements were performed in far geometry, namely the activated foil was placed at a distance of 10 cm from the surface of the HPGe detector. Due to the half-life the ^{109}Pd and ^{198}Au radioisotopes are rather long, the counting time can be extended in order to obtain a good statistics. A typical γ-ray spectrum and its photo-peak used in the activity measurement of the irradiated palladium foil is shown in Fig. 2.

3. Data analysis

3.1. γ-ray spectra analysis

The γ-ray spectra were analyzed with the GammaVision software, version 5.10. The activities of the ^{116m}In, ^{198}Au and ^{109}Pd radio-nuclides were determined from the 1293.56 keV (84.8%), 411.80 keV (95.62%) and 88.04 keV (3.67%) γ-rays, respectively. These γ-rays are interference free, well separated and have relatively high intensity. Therefore, the γ-ray interference corrections were avoided. However, the counting losses due to the attenuation of the γ-rays were taken into account and corrections were made by using the following expression: $F_g = \mu t/(1-e^{-\mu t})$, where F_g is the attenuation factor, μ is the linear attenuation coefficient and t is the sample thickness. The computed attenuation factor for the 88.04 keV γ-ray in Pd foils is 0.945, and that for the 411.80 keV of Au is 0.994, respectively. The attenuation factor for the 1293.56 keV γ-ray in indium (In) foil is 0.999, and it is not necessary to take into account. The measured activity was corrected to zero attenuation by dividing with factor *Fg*.

3.2. Determination of the thermal neutron cross-section

The details of the experimental procedure for the determination of the thermal neutron capture cross-section and resonance integral were described elsewhere [23–25]. The thermal neutron capture cross-section of the ^{108}Pd(n,γ)^{109}Pd reaction, $\sigma_{0,Pd}$ was determined relative to that of the ^{197}Au(n,γ)^{198}Au reaction based on the measured reaction rates of the ^{109}Pd and ^{198}Au radionuclide as follows [23–25]:

$$\sigma_{0,Pd} = \sigma_{0,Au} \times \frac{R_{Pd} - F_{Pd,Cd} R_{Pd,Cd}}{R_{Au} - F_{Au,Cd} R_{Au,Cd}} \times \frac{G_{th,Au}}{G_{th,Pd}} \times \frac{g_{Au}}{g_{Pd}}, \quad (1)$$

where $\sigma_{0,Au}$ is the thermal neutron capture cross-section of the ^{197}Au(n,γ)^{198}Au reaction, R_x and $R_{x,Cd}$ are reaction rates per atom for the sample x (Pd or Au) irradiated with and without the Cd cover, respectively. $F_{x,cd}$ is the cadmium correction factor, where $F_{pd,Cd} = 1.000$ [26] and $F_{Au,Cd} = 1.009$ [26]. The Westcott factor for the ^{108}Pd(n,γ)^{109}Pd reaction, $g_{Pd} = 1.0096$ [27], and for the ^{197}Au(n,γ)^{198}Au reaction, $g_{Au} = 1.0054$ [27]. $G_{th,x}$ is the thermal neutron self-shielding factor for x (Pd or Au) sample, which were determined by both the analytical method [28,29] and by the Monte Carlo simulation. The Monte Carlo simulation will be presented in the next Subsection 3.4.

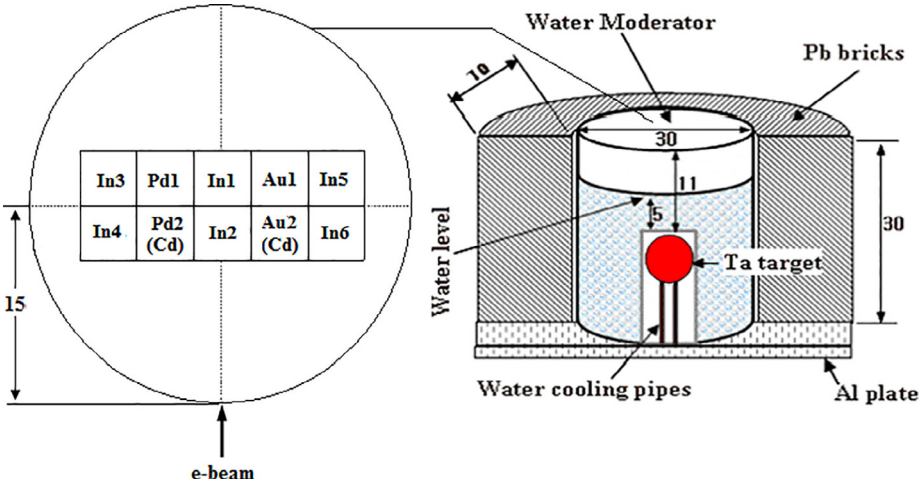

Fig. 1. Cross-sectional view of the Pohang electron linac based neutron source and arrangement of the activation foils.