열중성자에 대한 프라세오디뮴의 중성자포획확률에 대한 연구

Study on Neutron Capture Probability of Praseodymium at Thermal Neutron Energy

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요 약

기존의 $^{141}Pr(n,\gamma)^{142}Pr$ 반응에 대한 열중성자포획단면적 결과들은 여러 종류의 값들이 보고 되어 있다. 본 연구에서는 이상적인 중성자속을 가지는 교토원자로실험소의 중수열중성자장치를 이용하여 방사화 방법을 통해 열중성자포획 단면적을 보다 정밀하게 측정하였다. 시료에 입사되는 열 중성자속은 $^{197}Au(n,\gamma)^{198}Au$ 반응을 통하여 측정되었다. 측정된 결과는 기존의 측정 결과 및 JENDL-3.2, ENDF/ B-VI, JEF-2.2의 평가치들과 비교하였다.

Abstract

The thermal neutron capture cross-section (at 2,200 m/s value) of the $^{141}Pr(n,\gamma)^{142}Pr$ reaction was measured by an activation method by using the heavy water (D_2O) thermal neutron facility at the KUR(Kyoto University Reactor). The thermal neutron flux used in this experiment was monitored with the $^{197}Au(n,\gamma)^{198}Au$ standard cross-section.

The previous results and the evaluated data of JENDL-3.2, ENDF/B-VI, and JEF-2.2 were in good agreement with the current result.

I. Introduction

Praseodymium (^{141}Pr) is a kind of fission products (FPs) which are produced in Light Water Reactors (LWRs) and Liquid-Metal- Cooled Fast Breeders Reactors (LMFBRs). The cumulative fission yields of the thermal neutron-induced fission of ^{235}U and the fast neutron-induced fission of ^{239}Pu are 5.8 and 5.2 %, respectively [1]. These yield values are pretty large in the fission products and the ^{141}Pr nuclide may be apt to accumulate in the spent fuels. Therefore, accurate measurement of the nuclear data, especially a neutron capture cross-section, is important for the assessment of the reactor safety and for the investigation of high-burn-up core characteristics [2],[3]. According to the evaluated data in ENDF/B-VI [4], the capture cross-sections in the thermal and the keV energy regions are 11.5 b and about 0.1 b, respectively. Since the ^{141}Pr nuclide has the neutron magic number (N=82) and excess protons on the 2d5/2 sub-shell,

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the capture cross-section may not be so large as expected. However, the accurate nuclear data of the ^{141}Pr is thought to be needed for evaluating the nuclear transmutation performance in the reactors. Moreover, in keV energy range, the capture cross-section is also important for the study on the s-process for nucleosynthesis in stars [5]. Concerning the thermal neutron cross-sections (at 2,200 m/s value) for the ^{141}Pr(n,γ)^{142}Pr nuclear reaction, several measurements have been made with reactors. The measured results are shown in Table I [4],[6-16]. Pomerance [8] and Cummins [9] measured the data with the pile oscillator method. The data of Seren et al. [10], Lyon [11], Zimmerman et al. [12], Fehr and Hansen [13], Heft [14], and Kern et al. [15] were measured by the activation method. Their data are rather old and seem to be discrepant from each other. The evaluated data of JENDL-3.2, ENDF/B-VI, and JEF-2.2 are in good agreement, and those by Mughabghab et al. [16] are about 11.5 b at 0.0253 eV. This value is also in good agreement with three evaluated data.

As mentioned above, the precision of the given experimental data is not always satisfactory in quality and quantity. The thermal neutron capture cross-section was also measured by the foil activation method by using the heavy water thermal neutron facility of the Kyoto University Reactor (KUR) at the KURRI [17]. The reactor can produce very good thermal neutrons than other facilities in the world. The thermal neutron flux used in this measurement was monitored with a gold (Au) foil and the ^{197}Au(n,γ)^{198}Au standard cross-section. The TOF (Time-of-Flight) measurement was normalized to this activation data, and the result was compared with other previous experimental data and the evaluated cross- sections of JENDL-3.2, ENDF/B-VI, and JEF-2.2.

II. Experiment by Activation Method

1. Experimental Arrangement

The thermal neutron cross-section for the ^{141}Pr(n,γ)^{142}Pr reaction was measured by the activation method by using a heavy water thermal neutron facility at Kyoto University. The KUR is a highly enriched uranium-fueled research reactor, which is light water-moderated, and its nominal power is 5 MW [17]. Beside the core, there is a irradiation facility with a heavy water tank of 1.4 m in length. Outside the heavy water tank, there is an irradiation room of about 2.4 × 2.4 × 2.4 m^3 surrounded by 90 cm thick heavy concrete shields, as shown in Fig. 1. The leakage neutrons from the heavy water tank can be used as a thermal neutron source of plane-type in a large space. Sakurai et al. [18] obtained

Table I. Thermal neutron capture cross-sections for the ^{141}Pr(n,γ)^{142}Pr reaction

Reference	Cross-section (b)	Experimental method
L. Seren et al. (1947) [10]	10.1±2.02	Activation
H. Pomerance (1951) [8]	11.2±0.6	Pile oscillator
J. D. Cummins (1957) [9]	11.6±0.2	Pile oscillator
E. Fehr and E. Hansen (1960) [13]	9.2±1.0	Activation
W. S. Lyon (1960) [11]	10.9	Activation
J. Kern et al. (1967) [15]	3.7±0.5	Activation
R. L. Zimmerman et al. (1967) [12]	11.5±1.0	Activation
R. E. Heft (1978) [14]	8.36±0.1	Activation
S. F. Mughabghab et al. (1981) [16]	11.5±0.3	-
JENDL-3.2 (1998) [6]	11.500	-
ENDF/B-VI (1991) [4]	11.5	-
JEF-2.2 (1994) [7]	11.5	-

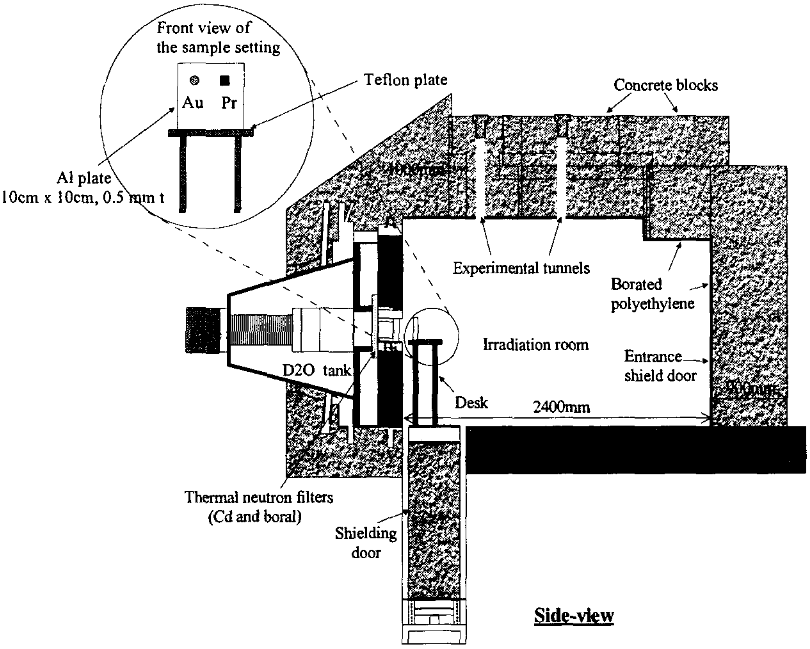

Fig. 1. The irradiation facility of KUR with a heavy water tank (D2O) at KURRI.

the neutron spectrum at the facility, whose spectrum showed a good Maxwellian distribution having a neutron temperature of 40°C. The Cd-ratio measured by Au foils without and with a Cd-cover of 0.7 mm in thickness was more than 780, and epithermal neutrons were almost negligible at the facility.

neutron flux. These foils were attached to an aluminum holder with adhesive tape and set at about 10 cm distance from the Bi surface, as shown in Fig. 1. The characteristics of the activation samples used in this current measurement are summarized in Table II.

Table II. Physical parameters of the samples used in our measurement

Samples	Physical form	Purity	Thickness			Size
–	–	(%)	(g/cm^2)	(atoms/kb)	(mm)	–
Pr	Foil	99.9	0.088	0.376	0.1	1.0×1.0 cm^2
Au	Foil	99.9	0.047	0.144	0.05	0.95 cm in diam.

2. Sample Preparation

A ^{141}Pr foil of 1.0 × 1.0 cm^2 in area, 0.1 mm in thickness and 99.9 % in chemical purity, was used for the thermal neutron capture cross-section measurement. A ^{197}Au foil of 0.95 cm in diameter, 50 m in thickness and 99.9 % in chemical purity, was used for monitoring the thermal

3. Measurement

For the thermal neutron cross-section measurement, a sample set of ^{141}Pr and ^{197}Au foils was attached to an aluminum holder and irradiated for 30 min. at the thermal neutron facility, as shown in Fig. 1. The ^{197}Au foil was used for the measurement of thermal neutron flux. The neutron flux at the irradiation position was about 1.5 × 109 n/cm2s under the 5MW operation of the KUR.

The activities induced from the irradiated samples of ^{142}Pr and Au were measured by using a HPGe detector which was well-shielded to natural background with a lead housing. Each of the samples was set at a distance of 5 cm from the detector case. The photo-peak efficiencies of the detector were experimentally calibrated with mixed γ-ray standard sources. The γ-ray energies and intensities [19] used for the current data processing are shown in Table III. The Westcott's g-factors for both of ^{141}Pr and ^{197}Au are also given in Table III [20].

$g_x(T_n)$ is the Westcott's g-factor, ϵ_x is the detection efficiency, N_x is the number of atoms for the relevant reaction, and ϕ_x is the neutron flux.

In this measurement, the thermal neutron cross-section for the Pr(n,γ) reaction at a neutron energy of 0.0253 eV (corresponding to a velocity of 2,200 m/s) was measured relative to that for the ^{197}Au(n,γ)^{198}Au reaction as a standard, by rewriting the above relations as follows: where pr and au denote the parameters for ^{141}Pr and ^{197}Au, respectively. The Westcott's g-factor of ^{197}Au was referred to as the result obtained by Gryntakis and Kim [20]

$$\sigma_{pr}(\nu_0) = \frac{\epsilon_{au}}{\epsilon_{pr}} \cdot \frac{R_{pr}}{R_{au}} \cdot \frac{N_{au}}{N_{pr}} \cdot \frac{S_{au}}{S_{pr}} \cdot \frac{g_{au}(T_n)}{g_{pr}(T_n)} \cdot \sigma_{au}(\nu_0) \tag{3}$$

Table III. Nuclear data*of ^{141}Pr and ^{197}Au used for the current measurement.

Isotope	Half life	-ray energy (MeV)	-ray intensity (%)	Westcott's g-factor**
^{141}Pr	–	–	–	1.0053
^{142}Pr	19.13 hours	1.576	3.70	–
^{197}Au	–	–	–	1.0399
^{198}Au	2.694 days	0.412	95.5	–

* Referred to Browne and Firestone [19], ** Referred to Gryntakis and Kim [20]

4. Data Analysis

The thermal neutron capture cross-section σ_S averaged over the Maxwellian distribution spectrum is defined as [17]

$$\sigma_x = \frac{\sigma_x(v_0)}{1.128} S_x g_x(T_n)\sqrt{\frac{T_o}{T_n}} \tag{1}$$

and the measured reaction rate is given in the following relation:

$$R_x = \epsilon_x N_x \phi_x, \quad (x = pr, au) \tag{2}$$

where ν_0=2,200 m/s, T_0=293.6 K, T_n is the neutron temperature, S_x is the self-shielding correction factor, and the factor of ^{141}Pr was analytically calculated. The thermal neutron cross-section of the ^{141}Pr(n,γ)^{142}Pr reaction was obtained relative to the well-known thermal neutron cross-section (98.65 ± 0.09 b) for the ^{197}Au(n,γ)^{198}Au reaction [16].

5. Corrections and Uncertainties

The correction factor of the neutron self-shielding and/or neutron multiple scattering effects for the ^{141}Pr foil was calculated by the MCNP code [21] and the result was S_{pr}= 0.999 at 0.0253 eV. The factor for the ^{197}Au foil was S_{au}= 0.958 in our calculation. Parallel neutron beam from the thermal neutron facility strikes the irradiation foils, and the calculation procedure is almost the same as that for the measurement by the linac TOF method. The experimental uncertainties for the current measurement are summarized in Table IV. The uncertainties for the current measurement are from 2.98 to 3.04 %, which are mainly due to the statistical error (0.8 ± 1.0 %), the γ-ray detection efficiency of the HPGe detector (2.5 %), and the number of atoms in each sample (1.3 %).

IV. Result and Discussion

We measured the thermal neutron cross-section of the ^{141}Pr(n,γ)^{142}Pr reaction relative to the ^{197}Au(n,γ)^{198}Au standard cross-section at 0.0253 eV, using the thermal neutron facility of the KUR and the activation method. The current measurement obtained is 11.6 ± 0.35 b. Comparing with other previous results in Fig. 2, the results of Seren et

Table IV. Experimental uncertainties in the current measurement.

Reasons of Uncertainties Experimental Error	(%)
Statistical error	0.8 ~1.0
-ray detection efficiency of the HPGe	2.5
Number of atoms	1.3
Geometrical factor for irradiation	0.1
Correction for the neutron self-shielding and/or neutron scattering effects	0.5
Reference cross section value for the ^{197}Au(n,) reaction	0.2
Total uncertainty	2.98~3.04

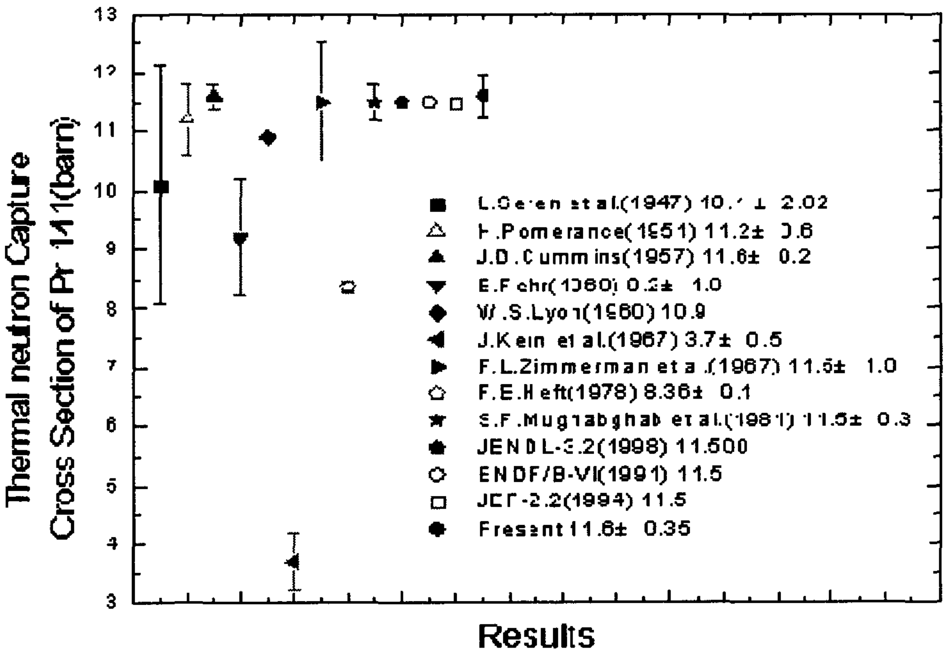

Fig. 2. Result

al. [10], Pomerance [8], Cummins [9], and Zimmerman et al. [12] seem to be in general agreement with the current measurement within the experimental error, although the data by Lyon [11], Fehr and Hansen [13], and Heft [14] are lower by 6.0 % and 28 % than the measurement, respectively. Especially, the data by Kern [15] is pretty lower by 68 %. The evaluated data of Mughabghab et al. [16], JENDL-3.2, ENDF/B-VI, and JEF-2.2 are in good agreement with the current value within the experimental uncertainty.

V. Conclusion

The thermal neutron cross-section of the $^{141}Pr(n,\gamma)^{142}Pr$ reaction was also measured with the activation method by using the heavy water thermal neutron facility of the KUR. The obtained result is 11.6 ± 0.35 b. Most of the previous measurements are in general agreement with the current measurement. However, the data by Fehr and Hansen, Lyon, and Heft are lower by 6.0 ~ 28 % than the current value. The thermal neutron cross-sections evaluated in JENDL-3.2, ENDF/B-VI, and JEF-2.2 and by Mughabghab et al. are in good agreement with the current measurement.

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