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Thermal neutron capture cross section and resonance integral of the ^{139}La(n,γ)^{140}La reaction

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ABSTRACT

The thermal neutron capture cross section (σ_0) and resonance integral cross section (I_0) of the ^{139}La(n,γ)^{140}La reaction have been measured relative to that of the ^{197}Au(n,γ)^{198}Au reaction by means of the activation method. High-purity natural lanthanum and gold foils were exposed to pulsed neutrons at the Pohang neutron facility. One set of foils was irradiated directly and a second set of foils was shielded with a cadmium cover of 0.5 mm thickness. The induced activities in the activated foils were measured by a γ-ray spectrometer based on a calibrated high purity germanium (HPGe) detector. In order to improve the accuracy of the experimental results the epithermal neutron spectrum shape factor (α) was determined, and the corrections for the thermal neutron self-shielding (G_{th}), the resonance neutron self-shielding (G_{epi}), the γ-ray attenuation (F_g) and the γ-ray coincidence summing effects were made. The thermal neutron cross-section for the ^{139}La(n,γ)^{140}La reaction has been determined to be 9.16 ± 0.36 barn, relative to the reference value of 98.65 ± 0.09 barn for the ^{197}Au(n,γ)^{198}Au reaction. By assuming the cadmium cut-off energy of 0.55 eV, the resonance integral cross section for the ^{139}La(n,γ)^{140}La reaction is 11.64 ± 0.69 barn, which is determined relative to the reference value of 1550 ± 28 barn for the ^{197}Au(n,γ)^{198}Au reaction. The measured results are compared with literature values and discussed.

1. Introduction

Natural lanthanum (^{nat}La) consists of the stable isotope ^{139}La with abundance of 99.9119(71)% and the radioactive isotope ^{138}La (half-life of $1.02(1) \times 10^{11}$ years) with abundance of 0.08881(71)%. The ^{139}La with $N = 82$ is known as the neutron magic isotope, which represents a promising s- and r-process indicator in stellar nucleosynthesis [1,2]. The ^{139}La is also an abundant fission product from the decay chain ^{139}Xe → ^{139}Cs → ^{139}Ba → ^{139}La in thermal neutron fission of ^{235}U [3], therefore it is widely used for neutron dosimetry in nuclear power plants [4] due to the half-life of ^{140}La is rather long ($T_{1/2} = 1.67855(12)$ d). In addition, the radioactive isotope ^{140}La is used as radiotracer in industrial and environmental applications [5]. We measured the thermal neutron capture cross section and the resonance integral of the ^{139}La(n,γ)^{140}La reaction relative to the reference values of the ^{197}Au(n,γ)^{198}Au reaction by using the activation method at the Pohang neutron facility (PNF) based on the 100-MeV electron linac of the Pohang accelerator laboratory (PAL). Recently, several similar neutron activation measurements were performed on this neutron facility [6,7].

There are 14 experimental [8–21] and 9 evaluated [22–30] data on the thermal neutron capture cross sections, and 10 experimental [10,13,15,31–37] and 8 evaluated [22–25,27–30] data on the resonance integral for the ^{139}La(n,γ)^{140}La reaction, respectively. The experimental thermal neutron cross sections vary from 8.1 [16] to 9.5 [15] barn, differ by about 17%, and the resonance integral data vary from 10.8 [36] to 17.1 [33] barn, differ by about 58%, respectively. Although the maximum deviation among the resonance integral is large, however most of them are in the range of 10.8–12.5 barn (differ by about 16%).

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The present work was undertaken with the aim to provide additional thermal neutron capture cross section and the resonance integral of the ^{139}La(n,γ)^{140}La reaction for comparison. In order to improve the accuracy of the experimental results, we corrected for the thermal neutron self-shielding (G_{th}) and the resonance neutron self-shielding (G_{epi}) effects, the γ-ray attenuation (F_g), and the γ-ray coincidence summing (F_{sum}) effects. Since the neutron field used in the sample activation is appropriated by the $1/E^{1+\alpha}$ distribution, there is remarkable effect in the final result due to the magnitude of the spectrum shape factor (α). We have also taken into account the effect of the spectrum shape factor (α) in the epithermal neutron spectrum in order to determine the resonance integral with high accuracy.

2. Experimental procedure

2.1. Neutron facility

The experiment was carried out at the PNF based on the 100-MeV electron linac of the Pohang accelerator laboratory. The characteristics of the PNF are described elsewhere [38–40], so only a general description is given here. The PNF consists of an electron linac, a photo-neutron target, and a 12-m long time-of-flight (TOF) path. The electron beam impinges upon a water-cooled tantalum target and produce bremsstrahlung radiation, which in turn generate photo-neutrons. The distributions of neutrons with and without water moderator were described elsewhere [41,42]. The photo-neutrons produced in the giant dipole resonance region consist of a large portion of evaporated neutrons and a small fraction of directly emitted neutrons which dominated at high energies. The neutrons produced in the Ta target without water moderator have a Maxwellian energy distribution with a nuclear temperature of 0.45 MeV. The estimated neutron yield per kW of beam power for electron energies above 50 MeV at the Ta target is about 1.9×10^{12} n/s [41], which is consistent with the calculated value based on Swanson's formula, $1.2 \times 10^{11}\ Z^{0.66}$, where Z is the atomic number of the target material [43]. The total neutron yield per kW of beam power was also measured by using the multiple-foil technique and found $(2.30 \pm 0.28) \times 10^{12}$ n/s [42]. The neutron energy spectrum with the water moderator is shifted to the lower energy region because of moderation by the water. In order to increase the thermal neutrons we have used water to a level of 3–5 cm above the Ta target surface [41]. In this experiment the water level was 5 cm above the target surface. The cadmium ratio defined as $CR = (R/R_{Cd})$, where R and R_{Cd} are reaction rates per atom for bare and Cd-covered isotope irradiation, were measured to be 2.96 ± 0.05 for ^{197}Au and 7.58 ± 0.09 for ^{139}La, respectively.

2.2. Sample preparation and irradiation

Two natural lanthanum metallic foils, 12×12 mm square and 0.1 mm in thickness, were prepared as the activation samples. In addition, we have also prepared two gold (Au) and six indium (In) metallic foils with the same size for comparator reactions and as neutron fluence monitors, respectively. The characteristics of these samples are given in Table 1.

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

Foils	Size (mm)	Thickness (mm)	Weight (g)	Purity (%)
La1	12 × 12	0.1	0.0955	99.9
La2	12 × 12	0.1	0.0981	99.9
Au1	12 × 12	0.1	0.3173	99.95
Au2	12 × 12	0.1	0.3082	99.95
In1	12 × 12	0.05	0.0505	99.99
In2	12 × 12	0.05	0.0558	99.99
In3	12 × 12	0.05	0.0556	99.99
In4	12 × 12	0.05	0.0534	99.99
In5	12 × 12	0.05	0.0565	99.99
In6	12 × 12	0.05	0.0569	99.99

The prepared La, Au, and In foils were irradiated simultaneously. The La and Au foils were irradiated with and without a Cd cover of 0.5 mm thickness. The neutron fluence exposed to La and Au foils was determined from the measured activities of the In monitors. For the neutron irradiations, the La, Au, and In foils were placed on the top of the water moderator as shown in Fig. 1, where La2(Cd) and Au2(Cd) denote the activation samples covered with a 0.5 mm Cd shield. The irradiation time was 140 min yielding enough the activities to be measured in a γ-ray counting system. The considered nuclear reactions ^{139}La(n,γ)^{140}La, ^{197}Au(n,γ)^{198}Au, and ^{115}In(n,γ)^{116m}In and their decay data used in calculations are given in Table 2 [44].

2.3. Activity measurement

The induced activities of the activated foils were measured by using an energy- and efficiency-calibrated high-purity germanium (HPGe) detector (ORTEC, GEM-20180-p) coupled to a PC-based 4 K channel analyzer. The photo-peak area was determined using the Gamma Vision, version 5.10. The energy resolution of the detector was 1.8 keV full width at half maximum (FWHM) at the 1332.5-keV peak of ^{60}Co. The detection efficiency is 20% at 1332.5 keV relative to a 3″ diameter × 3″ length NaI(Tl) detector. The absolute photopeak efficiencies and total efficiencies of the HPGe detector were measured with the calibrated γ-sources. The details of the detector efficiency calibrations were illustrated in Refs. [45,46].

For the activity measurement, the activated foil was attached to a plastic sample holder and it can be set at distances from 0.5 to 20 cm from the surface of the HPGe detector by 1 cm step. We first measured the activity of the indium foils after irradiation because the half-life of ^{116m}In is considerably shorter than that of the ^{140}La and ^{198}Au. Since the half-lives of ^{140}La and ^{198}Au are rather long, the measuring times for -ray spectra $v$aried from few 10 min to some hours depending on the statistics of the γ-ray peaks of interest. The induced activities in the ^{116m}In and ^{198}Au foils were determined by measuring the γ-rays of 1293.56 keV (84.4%) and 411.8 keV, respectively. The activities of the ^{140}La foils were determined by measuring the γ-rays at 328.76 keV (20.3%) and 487.02 keV (45.5%). The long counting distances were selected to reduce the counting losses due to true coincidence summing and pile-up effects. Usually, the dead-time of the data acquisition system was kept below 2%. The partial γ-ray spectrum from the La foil irradiated with moderated neutrons is given in Fig. 2.

3. Data analysis

3.1. Determination of the thermal neutron capture cross section

The thermal neutron capture cross section for the ^{139}La(n,γ)^{140}La reaction, $\sigma_{0,La}$, has been determined relative to that of the ^{197}Au(n,γ)^{198}Au reaction as follows [47]:

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

where $\sigma_{0,Au}$ is the thermal neutron cross section of the ^{197}Au(n,γ)^{198}Au reaction, R_x and $R_{x,Cd}$ are reaction rates per atom for bare and Cd-covered x (La or Au) isotope irradiation, respectively. The cadmium transmission correction factor, $F_{x,Cd}$ accounts for the difference in count rate for Cd covered and bare samples, and $G_{th,x}$ is the thermal neutron self-shielding factor for x sample. The Westcott correction factor, g_x accounts for the deviation of