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Measurement of thermal neutron cross-section and resonance integral for the ^{165}Ho(n,γ)^{166g}Ho reaction using electron linac-based neutron source

Van Do Nguyen [a], Duc Khue Pham [a], Tien Thanh Kim [a], Guinyun Kim [b,*], Manwoo Lee [b], Kyung Sook Kim [b], Heung-Sik Kang [c], Moo-Hyun Cho [c], In Soo Ko [c], Won Namkung [c]

[a] Institute of Physics, Vietnam Academy of Science and Technology, 10 Dao Tan, Hanoi, Viet Nam
[b] Department of Physics, Kyungpook National University, Daegu 702-701, Republic of Korea
[c] Pohang Accelerator Laboratory, Pohang University of Science and Technology, Pohang 790-784, Republic of Korea

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ABSTRACT

The thermal neutron cross-section and the resonance integral of the ^{165}Ho(n,γ)^{166g}Ho reaction have been measured by the activation method using a ^{197}Au(n,γ)^{198}Au monitor reaction as a single comparator. The high-purity natural Ho and Au foils with and without a cadmium shield case of 0.5 mm thickness were irradiated in a neutron field of the Pohang neutron facility. The induced activities in the activated foils were measured with a calibrated p-type high-purity Ge detector. The correction factors for the γ-ray attenuation (F_g), the thermal neutron self-shielding (G_{th}), the resonance neutron self-shielding (G_{epi}) effects, and the epithermal neutron spectrum shape factor (α) were taken into account. The thermal neutron cross-section for the ^{165}Ho(n,γ)^{166g}Ho reaction has been determined to be 59.7 ± 2.5 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 for the ^{165}Ho(n,γ)^{166g}Ho reaction is 671 ± 47 barn, which is determined relative to the reference value of 1550 ± 28 barn for the ^{197}Au(n,γ)^{198}Au reaction. The present results are, in general, good agreement with most of the previously reported data within uncertainty limits.

1. Introduction

The stable isotope ^{165}Ho has 100% natural abundance. This rare earth element can be used in nuclear reactors for nuclear control rods due to the thermal neutron cross-section and epithermal resonance integral for the ^{165}Ho(n,γ)^{166g}Ho reaction are rather high. In addition, ^{166g}Ho is one of the preferred radionuclides for nuclear medicine applications [1] because of its physical properties, which include high-energy β radiation ($E_{β1}$ = 1855 keV (51%), $E_{β2}$ = 1776 - keV (48%), and $E_{βav}$ = 666 keV), a short half-life (26.76 h), and decay to a stable daughter. It also emits low-intensity and low-energy γ-rays (80.57 keV, 6.56%) which are suitable for imaging by a gamma camera.

The ^{166g}Ho can be produced by either direct neutron capture of ^{165}Ho or by indirect nuclear reaction ^{164}Dy(n,γ)^{165}Dy(n,γ)^{166}-Dy(β$^-$)^{166g}Ho [1,2]. However, in practical applications, the production of ^{166g}Ho through ^{165}Ho(n,γ)^{166g}Ho reaction was widely used. Therefore, the knowledge of thermal neutron cross-section and resonance integral of the ^{165}Ho(n,γ)^{166g}Ho reaction would become important because the neutron activation cross-section data are needed for the production of ^{166g}Ho and may also used in other studies related to the interaction of neutrons with matters.

There are about 30 measurements and evaluations on the thermal neutron capture cross-sections and the resonance integrals for the ^{165}Ho(n,γ)^{166g}Ho reaction in the literature [3–31] from 1947 to 2009. However, the measured thermal neutron cross-sections for the ^{165}Ho(n,γ)^{166g}Ho reaction are varied from 58 [8] to 67 barn [27]. This fact shows that, there are still large deviations (15.5%) among the experimental results. The measured resonance integrals for the ^{165}Ho(n,γ)^{166g}Ho reaction are varied from 600 [25] to 830 barn [23]. The difference between the lowest and the highest values of the resonance integral found in literature is 33.8%. Therefore, it is necessary to measure more new data for better comparison and evaluation.

We measured the thermal neutron cross-section and resonance integral of the ^{165}Ho(n,γ)^{166g}Ho reaction by using the well known activation method at the Pohang Neutron Facility (PNF) based on the 65-MeV electron linear accelerator (linac). There are few measurements similar to this work done at PNF [32]. The thermal neutron cross-section and the resonance integral for the ^{165}Ho(n,γ)^{166g}Ho reaction were determined relative to the reference values of the ^{197}Au(n,γ)^{198}Au reaction. In this experiment the necessary correction factors for γ-ray attenuation (F_g), thermal

* Corresponding author. Tel.: + 82 53 950 5320; fax: +82 53 955 5356.
E-mail address: gnkim@knu.ac.kr (G. Kim).

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neutron self-shielding (G_{th}), and resonance neutron self-shielding (G_{epi}) effects were taken into account. In addition, the neutron field used in sample activations, in general, approximately follow a $1/E^{1+\alpha}$ distribution, thus giving rise to the remarkable effect in the final result due to the magnitude of the spectrum shape factor (α). Therefore, in order to improve the accuracy of the resonance integral result, the effect of the spectrum shape factor (α) in epithermal neutron spectrum was also taken into account. The present results are compared with the existing experimental data and the evaluated values.

2. Experimental procedure

2.1. Neutron source

Neutrons used in this experiment were produced from the Pohang Neutron Facility (PNF) based on the pulsed electron linac of the Pohang Accelerator Laboratory (PAL), Korea [33]. The characteristics of the PNF are described elsewhere [34–38], so only a general description is given here. It consists of an electron linac, a photo-neutron target, and a 12-m long time-of-flight (TOF) path. The photo-neutron target was composed of ten Ta plates with a diameter of 4.9 cm and an effective thickness of 7.4 cm. There was a 0.15 cm water gap between Ta plates in order to cool the target effectively. The housing of the target was made of titanium. The photo-neutron target was set in the center of a cylindrical water moderator. The water moderator made by an aluminum cylinder with a thickness of 0.5 cm, a diameter of 30 cm, and a height of 30 cm.

The distributions of neutrons with and without water moderator were described elsewhere [39,40]. 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 [39], 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 [41]. 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 [40]. The neutron energy spectrum with the water moderator is shifted to lower energy region because of the effect of moderation by water. To increase the thermal neutrons in this facility, we have used water to a level of 3–5 cm above the Ta target surface [39]. In this experiment the water level was 5 cm above the target surface.

2.2. Sample irradiation

High-purity (99.9%) natural Holmium foils, 10×10 mm rectangular shape and 0.025 mm in thickness, were used as the activation samples. The Au and In metallic foils were used as the comparator reactions and the neutron flux monitors, respectively. The characteristics of Ho, Au, and In foils are given in Table 1.

In order to measure the thermal neutron cross-section and the resonance integral for the ^{165}Ho(n,γ)^{166g}Ho reaction by activation method relative to the ^{197}Au(n,γ)^{198}Au reaction, the natural Ho and Au foils were irradiated with and without a Cd cover with a thickness of 0.5 mm. The neutron fluxes exposed to each sample during the irradiation were determined from activities of In monitors stacked alternatively between Ho and Au foils. The Ho, Au, and In foils were stacked on the sample holder, and the sample holder was placed on the upper surface of the water moderator as shown in Fig. 1, where Ho(Cd) and Au(Cd) denote the activation foil covered with a 0.5-mm thick Cd. The neutron flux exposed to each sample was extrapolated from the measured activities of In foils irradiated simultaneously with the foil samples. The cadmium ratio is defined by $CR = (R/R_{cd})$, where R and R_{cd} are reaction rates per atom for bare and Cd-covered isotope irradiation, respectively. The measured cadmium ratio for ^{197}Au was 2.76 ± 0.04, and that for ^{165}Ho was 2.18 ± 0.03, respectively.

Table 1
Characteristics of the activation foils: Ho, Au and In.

Foils	Size (mm)	Thickness (mm)	Weight (g)	Purity (%)
Ho1	10.0 × 10.0	0.025	0.0206	99.9
Ho2	10.0 × 10.0	0.025	0.0207	99.9
AuC	10.0 × 10.0	0.03	0.0467	99.95
AuD	10.0 × 10.0	0.03	0.0463	99.95
In13	10.0 × 10.0	0.05	0.0316	99.95
In14	10.0 × 10.0	0.05	0.0314	99.95
In15	10.0 × 10.0	0.05	0.0327	99.95
In43	10.0 × 10.0	0.05	0.0318	99.95
In44	10.0 × 10.0	0.05	0.0326	99.95
In45	10.0 × 10.0	0.05	0.0323	99.95

In this study, the irradiation time was 180 min. The main nuclear data together with their uncertainties given in parenthesis for the nuclear reactions considered such as ^{165}Ho(n,γ)^{166g}Ho, ^{197}Au(n,γ)^{198}Au, and ^{115}In(n,γ)^{116m}In are listed in Table 2 based on the table of isotopes [42].

2.3. Measurement of activity

After an irradiation and an appropriate waiting time, the irradiated foils were taken off, and the activities of ^{166g}Ho, ^{198}Au, and ^{116m}In were measured by using a high-resolution γ-ray spectrometer. The γ-ray spectrometer was a p-type coaxial CANBERRA high-purity germanium (HPGe) detector with a diameter of 59.2 mm and a thickness of 30 mm. The HPGe-detector was coupled to a computer based multi-channel analyzer card system, which can determine the photo-peak-area of γ-ray spectra by using the GENIE2000 computer program. The energy resolution of the detector was 1.80 keV full width at half maximum (FWHM) at the 1332.501 keV peak of ^{60}Co. The detection efficiency is 20% at 1332.501 keV relative to a 3″ diameter × 3″ length NaI(Tl) detector. The detection efficiency for the γ-ray spectrometer was calibrated with a set of standard γ-ray sources such as ^{241}Am (59.541 keV), ^{137}Cs (661.657 keV), ^{54}Mn (834.848 keV), ^{60}Co (1173.237 keV and 1332.501 keV), and ^{133}Ba (80.997 keV; 276.398 keV; 302.853 keV 356.017 and 383.815 keV). The measured detection efficiencies were fitted by the following function:

$$\ln \varepsilon = \sum_{n=0}^{5} a_n (\ln (E/E_0))^n \quad (1)$$

where ε is the detection efficiency, a_n represents the fitting parameters, and E is the energy of the photopeak, and $E_0 = 1$ keV. The detection efficiencies as a function of the photon energy measured at different distances between the gamma source and the surface of the detector were illustrated in Ref. [32].

The waiting and the measuring times were chosen based on the activity and the half-life of each radioactive isotope. In order to minimize the uncertainties caused by random coincidence and pile-up effects, we have chosen the appropriate distance between the sample and the detector for each measurement. Generally, the dead times were kept below 2% for all measurements. For this purpose, the measured foil was attached on plastic holder and can be set at a distance from 5 to 105 mm from the surface of the HPGe detector.