Annals of Nuclear Energy 63 (2014) 199–204

Contents lists available at ScienceDirect

Annals of Nuclear Energy

journal homepage: www.elsevier.com/locate/anucene

Measurement of thermal neutron cross section and resonance integral for the ^{158}Gd(n, γ)^{159}Gd reaction by using a ^{55}Mn monitor

Mustafa Karadag [a,*], M. Güray Budak [a], Haluk Yücel [b]

[a] Gazi University, Gazi Education Faculty, 06500 Teknikokullar-Ankara, Turkey
[b] Ankara University, Institute of Nuclear Sciences, 06100 Beşevler-Ankara, Turkey

ARTICLE INFO

Article history:
Received 15 May 2013
Received in revised form 10 July 2013
Accepted 13 July 2013
Available online 23 August 2013

Keywords:
Nuclear reaction
Activation method
Cadmium ratio
Thermal neutron cross section
Resonance integral

ABSTRACT

The thermal neutron cross section and the resonance integral for the ^{158}Gd(n, γ)^{159}Gd reaction were measured by the activation method with using the ^{55}Mn(n, γ)^{56}Mn monitor reaction. The analytical grade MnO_2 and Gd_2O_3 were mixed separately with Al_2O_3 powder to reduce both thermal and epithermal neutron self-shielding effects. Then, they were irradiated with and without a cadmium shield box in an isotropic neutron field provided by three ^{241}Am–Be neutron sources. The induced activities in the samples were measured by a p-type HPGe detector. For the irradiated samples, epithermal neutron spectrum-shape factor (α), correction factors required for thermal and epithermal neutron self-shielding and for the measured γ-rays, self-absorption and true coincidence summing effects were taken into account. The thermal neutron cross section was measured to be 2.15 ± 0.14 *b* for the ^{158}Gd(n, γ)^{159}Gd reaction. The previously reported experimental data for the thermal neutron cross section are distributed from 0.9 to 3.89 *b*. According to the definition of Cd cut-off energy at 0.55 eV, the resonance integral cross section was measured as 75.3 ± 7.0 *b*. The present resonance integral value agrees with some previously reported experimental and evaluated values for the ^{158}Gd(n, γ)^{159}Gd reaction, e.g. 80 *b* measured by Van DerLinden et al. (1974), 72.06 *b* evaluated by JENDL-4.0 (Shibata et al., 2011), 73 *b* reported by Mughabghab (2006) and Holden (1999).

1. Introduction

Gadolinium-159 (^{159}Gd) is a beta emitting therapeutic radionuclide with half-life of 18.479 h that emits a beta with maximum energy of 970.5 keV and a few gamma radiations (its main γ-ray energy: 363.543 keV), and delivers a higher radiation dose rate suitable for therapeutic applications in nuclear medicine, such as alleviating bone pain due to metastases, synovectomy, and tumor therapy (Soares et al., 2010, 2011). ^{159}Gd can be produced via the ^{158}Gd(n, γ)^{159}Gd reaction by thermal and epithermal neutrons irradiations of the target isotope ^{158}Gd, which is one of the six stable isotopes of the rare-earth gadolinium element. Hence, the thermal neutron cross section (σ_0) and the resonance integral cross section (I_0) of the ^{158}Gd(n, γ)^{159}Gd reaction are great importance not only for therapeutic radioisotope production of ^{159}Gd but also for studies concerning interactions of neutrons with matter and other fundamental nuclear research.

In surveying of literature the existing experimental values for σ_0 are 2.44 *b* by Heft (1978), 2.78 *b* by Mangal and Gill (1963), 3.89 *b* by Lyon (1960), 1.1 *b* by Butement (1949) and 0.9 *b* by Seren et al. (1947). Therefore, the discrepancies among them are up to 333%. Similarly, evaluated data for σ_0 range from 1.5 to 3.5 *b*. For the resonance integral value I_0, experimental valuesexisted in the literature are 94 *b* by Heft (1978), 127 *b* by Steinnes (1975), 80 *b* by Van Der Linden et al. (1974) and 84 *b* by Steinnes (1972). The discrepancies among these values are up to 59%. Also, the evaluated data for I_0 are distributed from 60.5 to 97.9 *b*. It seems that there has not been a sufficient consistency among σ_0 or I_0 values. However, the most recent experimental study to determine σ_0 andI_0 values was performed by Heft (1978) in 1978. Since then, a new experimental work for determination of them has not been performed with the state-of-the-art modern neutron metrology (EXFOR, 2013). Therefore, the aim of the present work was to carry out a new experiment with the state-of-the-art modern neutron metrology in order to clarify the existing differences in the σ_0 and I_0 values for the ^{158}Gd(n, γ)^{159}Gd reaction by employing Cd-ratio method with use of the ^{55}Mn(n, γ)^{56}Mn reaction as a single reference monitor. Hence, ^{55}Mn has been considered to be a convenient alternative monitor with good resonance parameters for theresonance integral determination in which most of the resonance captures occur at relatively higher neutron energy region at about 337 eV, which is quite far from 1/*v* region. Other advantages of ^{55}Mn as a monitor isotope were explicated previously (i.e., Karadag et al., 2003; Karadag and Yücel, 2004, 2005, 2008; Yücel

* Corresponding author. Tel.: +90 (312) 2028265; fax: +90 (312) 2228483.
E-mail address: mkaradag@gazi.edu.tr (M. Karadag).

0306-4549/$ - see front matter © 2013 Elsevier Ltd. All rights reserved.
http://dx.doi.org/10.1016/j.anucene.2013.07.015

and Karadag, 2005; Karadag et al., 2007; Yücel et al., 2007; Karadag, 2013a, 2013b).

2. Experimental

2.1. Samples

In this study, the powders of Gd_2O_3 and MnO_2 having high purities (>99.99%) obtained from Aldrich Inc. were used as samples. In order to minimize errors due to both thermal and epithermal neutron self-shielding effects, they were mixed independently from each other with sufficient amounts of Al_2O_3 powder because Al and O elements both have the lower neutron absorption cross section. The percentages of dilutions were experimentally determined to obtain minimum neutron self-absorptions in the samples. Then, the diluted samples of (3.2% Gd_2O_3 + 96.8% Al_2O_3) and (3.4% MnO_2 + 96.6% Al_2O_3) were filled in small polystyrene tubes with height/diameter ≈ 2 and wall thickness of 1 mm. Ten diluted Gd-samples having each mass of about 0.8 g were individually prepared. Five of them were used for Cd-covered irradiation and the other five samples were used for obtaining bare irradiation data. The same sample preparation procedure was employed for ten Mn-samples.

2.2. Irradiation and activity measurements

The sample irradiations were carried out with and without a cadmium (Cd) box, which is a cylinder box with a 1 mm wall thickness. The irradiator used in this work has an isotropic neutron field provided by three ^{241}Am–Be isotopic neutron sources each of which having 592 GBq activity, immersed in paraffin moderator shielded with lead bricks. The geometrical configuration of the neutron sources and the irradiation holes have been previously described in detail (see Karadag et al., 2003; Yücel and Karadag, 2004; Yücel et al., 2007). The effect of thermal flux depression at the sample irradiation position could be neglected, since the irradiation hole has a very large volume compared to the sample volume. At the sample irradiationposition, thermal and epithermal neutron fluxes were measured to be $(1.5 \pm 0.2) \times 10^4$ and $(1.4 \pm 0.1) \times 10^3$ -neutrons/cm^2 s respectively, and the epithermal neutron spectrum shape factor (α) was experimentally determined as 0.083 ± 0.016 in the previous work (Yücel and Karadag, 2004). The irradiation position was the same for all samples. The irradiation times for the (n, γ) reactions were chosen about 55–90 h for ^{158}Gd and about 7–13 h for ^{55}Mn yielding enough activity to be measured in the γ-ray counting system. After the irradiations, the suitable waiting-times varying up to 20 min were employed to minimize dead time losses and eliminate the possible contributions of 843.76 keV γ-ray by minor activity of ^{27}Mg (half-life of 9.458 m), which may be produced by the ^{27}Al(n, p)^{27}Mg reaction.

The samples were measured by a γ-ray counting system equipped with a coaxial p-type HPGe detector (Canberra GC11021) with a measured relative efficiency of 120.8%, an energy resolution of 1.95 keV, and a peak-to-Compton ratio of 85.7:1 at 1332.5 keV of ^{60}Co. The detector was connected to a digital spectrum analyzer with a full featured 16 K ADC/MCA analyzer based on digital signal processing operating through a Genie-2000 gamma spectroscopy software. The samples were counted at a distance of 10 cm from the detector in order to keep possible true coincidence effects (TCS) at a reasonable low level. For the fixed counting geometry, the gamma detection efficiency for the present HPGe detector was determined using a multinuclide standard in sand matrix (density: 1.7 g cm^{-3}) spiked with ^{241}Am, ^{109}Cd, ^{57}Co, ^{123m}Te, ^{51}Cr, ^{113}Sn, ^{85}Sr, ^{137}Cs, ^{60}Co, and ^{88}Y radionuclides, obtained from Isotope Products Laboratories Inc., traceable to National Institute of Standards and Technology (NIST). The counting periods predetermined for each measurement were high enough to ensure good statistical quality of data, so they varied between 2 and 24 h, depending on mainly the activity level of ^{56}Mn or ^{159}Gd products in the samples. Each spectrum was collected in the live-time mode. Dead times were typically less than about 0.1%. Background measurements were subtracted from the sample spectra.

3. Data evaluation procedure

3.1. Thermal neutron cross section

The thermal neutron cross section ($\sigma_{0,Gd}$) for the ^{158}Gd(n, γ)^{159}Gd reaction was determined relative to the reference value, $\sigma_{0,Mn}$ = 13.3 ± 0.1 *b* of the ^{55}Mn(n, γ)^{56}Mn reaction as follows:

$$\sigma_{0,Gd} = \frac{(r - r_{Cd})_{Gd}}{(r - r_{Cd})_{Mn}} \times \left(\frac{G_{th,Mn}}{G_{th,Gd}}\right) \times \left(\frac{g(20\,^{\circ}C)_{Mn}}{g(20\,^{\circ}C)_{Gd}}\right) \times \sigma_{0,Mn} \quad (1)$$

where Gd denotes the ^{158}Gd(n, γ)^{159}Gd reaction being investigated and Mn denotes the ^{55}Mn(n, γ)^{56}Mn monitor reaction. G_{th} is thermal neutron self-shielding factor. Since the Westcott correction factors, $g(20\,^{\circ}C)$ are 1.0009 for ^{158}Gd and 1.0003 for ^{55}Mn isotopes (Mughabghab, 2003), these isotopes can be considered a good 1/*v*-law behavior. Since the cadmium transmission factors (F_{Cd}) for ^{55}Mn and ^{158}Gd are unity (El Nimr et al., 1981), they may not be introduced into Eq. (1). The reaction rates per target atom, r and r_{Cd} are for bare and Cd-covered isotope irradiations, respectively. They have been determined for ^{56}Mn and ^{159}Gd isotopes by the following:

$$r = \frac{A_{sp}^{-} \cdot M \cdot F_g \cdot F_{coi}}{\theta \cdot N_A \cdot \gamma \cdot \varepsilon_p} \text{ and } r_{Cd} = \frac{A_{sp}^{+} \cdot M \cdot F_g \cdot F_{coi}}{\theta \cdot N_A \cdot \gamma \cdot \varepsilon_p} \quad (2)$$

with

$$A_{sp}^{-} \text{ or } A_{sp}^{+} = \frac{N_p}{t_m \cdot w \cdot S \cdot D \cdot C} \quad (3)$$

where A_{sp}^{-} and A_{sp}^{+} are specific activities obtained after a bare and Cd-covered isotope irradiations, N_p is the net number of counts under the full-energy peak collected during measuring (live) time t_m, w is weight of irradiated element, S is saturation factor: $S = 1 - e^{-\lambda\, t_{irr}}$ with decay constant, λ and irradiation time t_{irr}, D is decay factor: $D = e^{-\lambda\, t_d}$ with decay time t_d, C is measurement factor: $C = (1 - e^{-\lambda\, t_r})/\lambda\, t_r$ correcting for decay during the measuring true time t_r, M is atomic weight, θ is isotopic abundance, N_A is the Avogadro's number, γ is absolute gamma ray emission probability, ε_p is full-energy peak efficiency, F_g is correction factor for gamma ray attenuation and F_{coi} is true coincidence summing factor.

For gamma ray self-attenuations in the samples of (3.2%Gd_2O_3 + 96.8%Al_2O_3) and (3.4%MnO_2 + 96.6%Al_2O_3), the correction factors (F_g) were calculated at a fixed geometry for a small sample cylinder which is coaxially positioned with the detector. In the calculations of F_g, the two methods were used. In the first one, the simple equation: $F_g = \mu x/(1 - e^{-\mu x})$ was used (ASTM, 2003). Where x is sample thickness (cm) and μ is the linear attenuation coefficient (cm^{-1}), which can be determined easily from the total mass attenuation coefficients, μ/ρ (cm^2 g^{-1}) taken from the XCOM-Photon Cross Section Database (Berger et al., 2010). In the second one, a commercial software-GESPECOR® (Ver. 4.2) was also used to determine F_g factors. Then, the averages of F_g factors were calculated and taken into account in the measurements. True coincidence summing effects (TCS) for the γ-ray counting geometry were also taken into account by using MCNP method built in GESPECOR® software (Sima et al., 2001). Gamma ray self-attenuation factors (F_g), true coincidence summing factors (F_{coi}) and other nuclear data used in the analyses are given in Table 1. However, it