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Measurement of thermal neutron cross section and resonance integral for (n,γ) reaction in ^{152}Sm

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Abstract

This study implies that ^{55}Mn(n,γ)^{55}Mn monitor reaction may be a convenient alternative comparator for the activation method and thus, it was used for the determination of thermal neutron cross section (TNX) and the resonance integral (RI) of the reaction ^{152}Sm(n,γ)^{153}Sm. The samples of MnO_2 and Sm_2O_3 diluted with Al_2O_3 powder were irradiated within and without a cylindrical 1 mm-Cd shield case in an isotropic neutron field obtained from the ^{241}Am–Be neutron sources. The γ-ray spectra from the irradiated samples were measured by high resolution γ-ray spectrometry with a calibrated n-type Ge detector. The correction factors for γ-ray attenuation, thermal neutron and resonance neutron self-shielding effects and epithermal neutron spectrum shape factor (α) were taken into account in the determinations. The thermal neutron cross section for ^{152}Sm(n,γ)^{153}Sm reaction has been determined to be 204.8 ± 7.9 b at 0.025 eV. This result has been obtained relative to the reference thermal neutron cross section value of 13.3 ± 0.1 b for the ^{55}Mn(n,γ)^{56}Mn reaction. For the TNX, most of the experimental data and evaluated one in JEFF-3.1, ENDF/B-VI, JENDL 3.3 and BROND 2.0, in general, agree well with the present result. The RI value for ^{152}Sm(n,γ)^{153}Sm reaction has also been determined to be 3038 ± 214 b, relative to the reference value of 14.0 ± 0.3 b for the ^{55}Mn(n,γ)^{56}Mn monitor reaction, using a $1/E^{1+\alpha}$ epithermal neutron spectrum and assuming Cd cut-off energy of 0.55 eV. In surveying literature, the existing experimental and evaluated data for the RI values are distributed from 1715 to 3462 b. However, when the Cd cut-off energy is defined as 0.55 eV, the present RI value agrees with some previously reported RI values, 3020 ± 163 b by Simonits et al., 3141 ± 157 by Van Der Linden et al., and 2962 ± 54 b by Kafala et al., within the limits of error.

1. Introduction

Samarium (Sm) is a widely used rare earth element in nuclear reactors as a neutron absorber because of high thermal and epithermal neutron cross sections of some Sm isotopes, and has also a great importance in nuclear medicine for therapeutic purposes because of suitable decay and dose parameters of some radioisotopes to be delivered to the patient. For example, while a stable isotope, ^{152}Sm is used as control material in nuclear reactors, its produced radioactive isotope, ^{153}Sm is used as one of the β^- emitting therapeutic radioisotope in nuclear medicine for tumor therapy and bone pain palliation due to its high local beta dose per disintegration and suitable half-life (46.5 h). The radioactive isotope, ^{153}Sm disintegrates by about 82% via beta transitions to excited levels and by about 18% to the ground state of ^{153}Eu. The levels are depopulated by photon emission with strong gamma radiations at 69.67 (4.85%) and 103.18 keV (29.8%), while γ-ray emission probabilities for other transitions with energies up to 760 keV are weak (Knapp et al., 1998; Schötzig et al., 1999). Additionally, ^{153}Sm has recently been used in insect ecology (insects behaviours, such as dispersal) as ingested marker of some insects and detected to using neutron activation analysis (NAA) because the sensitivity of its detection by NAA is high (Showler et al., 2006). In general, ^{153}Sm can be produced in a nuclear reactor from the (n,γ)

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reaction in ^{152}Sm. The knowledge of the thermal neutron cross section (TNX that is, σ_0) and resonance integral (RI that is, I_0) of the ^{152}Sm(n,γ)^{153}Sm reaction would become important because the neutron activation cross section data are used in the production of ^{153}Sm and may also used in other studies related to the interaction of neutrons with matter. In surveying literature, it is worth noting that more than 40 experiments have been performed to measure the TNX and RI of ^{152}Sm(n,γ)^{153}Sm reaction. But there are still large discrepancies among RI values for this reaction. It is known that the TNX and RI for ^{152}Sm(n,γ)^{153}Sm reaction have been measured by activation method using various neutron sources with high neutron densities (generally reactors, accelerator-based neutron sources) and using the well-known gold monitor as a standard. However, accurate monitoring of highly intense neutron fields is complicated by the occurrence of burn up effects with some monitors. In the particular case of gold and samarium activation, significant burn up of the reaction products (^{198}Au and ^{153}Sm) can be expected, owing to their high TNX or RI values (e.g., $I_0 = 1.55 \times 10^3$ b for ^{198}Au, and $I_0 \cong 3.0 \times 10^3$ b for ^{153}Sm). Hence, the obtained specific activities of the products are not proportional to the neutron flux. Since the RI value of the ^{152}Sm(n,γ)^{153}Sm reaction are relatively high, a radioactive neutron source such as ^{241}Am–Be having with relatively low neutron flux seems to be a convenient alternative for RI determination by the activation method using a different monitor, ^{55}Mn with the good resonance parameters. Therefore, in this work, the ^{55}Mn(n,γ)^{56}Mn monitor reaction has been used for the TNX and RI determinations in which most of the resonance captures occurs at relatively higher energy region (first principal resonance of ^{55}Mn is at 337 eV, and its effective resonance energy is 468 eV), which is quite far from $1/v$ region. Therefore, the comparison with ^{197}Au-monitor does not need to correct for the effect of non-ideality of the epithermal neutron spectrum shape on the result for the nuclide, ^{153}Sm only if the epithermal neutron spectrum has an ideal-$1/E$ dependence on energy. But, it is the fact that the real epithermal neutron spectra in actual irradiation sites may deviate more or less from the ideal-$1/E$ distribution, and these deviating spectra approximately follow a $1/E^{1+\alpha}$ flux distribution. On the other hand, since the well separated principal resonance energy of ^{197}Au monitor lies in 4.9 eV, which is very close to $1/v$ thermal energy region, it is emphasized that the ^{55}Mn(n,γ)^{56}Mn reaction can also be chosen as a suitable comparator for RI measurements (Karadag et al., 2003; Yücel and Karadag, 2005). Thus, the motivation for the present measurements was the discrepancy among the RI values appeared in the literature survey, which can reach more than 50%. So, with the state of art modern neutron metrology in this study, it is aimed to carry out a new cross section measurement experiment in order to clarify the existing differences in RI values for the ^{152}Sm(n,γ)^{153}Sm reaction by using the activation method with cadmium ratios relative to ^{55}Mn(n,γ)^{56}Mn reaction.

2. Experimental

The samples were irradiated in an isotropic neutron field of the three ^{241}Am–Be isotopic neutron sources each having 592 GBq activity, immersed in paraffin moderator shielded with chevron type-lead bricks. The geometrical configuration of the neutron sources and the irradiation holes of this neutron irradiator installed at Ankara Nuclear Research and Training Center (ANRTC) has been previously described in detail, elsewhere (Karadag et al., 2003; Yücel and Karadag, 2004). This neutron irradiator was later moved to SNRTC in Sarayköy in Ankara, as of July 2005. According to the procedure described in the previous work (Yücel and Karadag, 2004), the equivalent 2200 m/s thermal and epithermal neutron fluxes at the sample irradiation position of the ^{241}Am–Be neutron irradiator have been measured to be $(1.5 \pm 0.2) \times 10^4$ and $(1.4 \pm 0.1) \times 10^3$ n cm^{-2} s^{-1}, respectively.

The oxide forms of the analytical grade samples to be activated were filled in the polystyrene tubes (height/diameter ≈ 2) with 1 mm wall thickness. The internal diameter of the sample tube is 5.0 mm. They were exposed to the neutrons in a fixed position in the irradiation hole of very large volume compared to the sample volume. Therefore, the effect of thermal flux depression at the irradiation site has been neglected. The extent of non-ideality of epithermal flux shape, α-factor at the sample irradiation position for the irradiation hole used of the neutron irradiator, was experimentally determined to be 0.083 ± 0.016 by dual monitor method using ^{98}Mo and ^{197}Au isotopes (Yücel and Karadag, 2004). In the present measurements, the oxide form of samarium was diluted sufficiently so as to minimize errors due to neutron self-shielding effect. For the sample dilution, the analytical grade purity and finely grained-Al_2O_3 powder is used by 96.6–99.0% in weight, because ^{27}Al isotope has the lower neutron absorption cross section. Each of the samples of MnO_2 and Sm_2O_3 was mixed independently each other with use of the sufficient amount of Al_2O_3 powder. The irradiations of samples were carried out with and without a cylindrical cadmium shield case. The irradiations for 12 samples for samarium element, which are individually prepared from the analytical grade stocks purchased from Aldrich Inc., were repeated five times that is, a set of 6, with and without Cd irradiation data for MnO_2 and Sm_2O_3 are obtained. The irradiation times for the (n,γ) reactions of ^{55}Mn and ^{152}Sm were chosen for a period of three to five half lives, yielding enough activity to be measured in a γ-ray counting system. The suitable waiting times (about 4 h) were employed to minimize dead time losses and eliminate the possible contributions of 104.32 keV γ-ray from ^{155}Sm (22.3 m) activity to the 103.18 keV peak of ^{153}Sm and 843.8 keV γ-ray from ^{27}Mg (9.45 m) activity to the 846.7 keV peak of ^{56}Mn.

The detector used in the measurements was an n-type high purity coaxial Ge (HPGe) with Be window, manufactured by Canberra Inc. The resolution of this detector was 1.80 keV for 1332.5 keV (^{60}Co) and 0.97 keV for 122 keV

(^{57}Co) and its measured relative efficiency was 22.6%. The detector was shielded by a 10 cm thick lead lined with copper sheets on all sides. The multi channel analyzer (MCA) system includes a full 8 K-channel MCA memory/ADC conversion gain for pulse height analysis, a high voltage power supply and a spectroscopy amplifier. The present HPGe detector was set up to collect 4096 channel spectra with a gain of about 0.48 keV/channel, using point gamma calibration sources. Each spectrum was collected in the live-time mode.

The gamma detection efficiency as a function of energy for the HPGe detector was determined using the powder radioactive standard containing a mixture of ^{241}Am, ^{109}Cd, ^{57}Co, $^{123\,m}$Te, ^{51}Cr, ^{113}Sn, ^{85}Sr, ^{137}Cs, ^{60}Co and ^{88}Y radionuclides, obtained from Isotope Products Laboratories Inc. traceable to NIST.

The counting times varied between 50 h and 120 h for Sm_2O_3 samples, depending on mainly the produced activity of Sm, and between 2.5 and 10 h for MnO_2 samples predetermined for each measurement were high enough to ensure good statistical quality of data. Dead times were typically less than 0.2%. Background measurements were subtracted from the sample spectra.

3. Results and discussion

The TNX (σ_0) and RI (I_0) for the reaction ^{152}Sm(n,γ)-^{153}Sm have been determined relative to that for the ^{55}Mn-(n,γ)^{56}Mn reaction, using the reaction rates of ^{153}Sm and ^{56}Mn with and without Cd. For the monitor reaction ^{55}Mn(n,γ)^{56}Mn, the reference values $\sigma_{0,Mn} = 13.3 \pm 0.1$ b and $I_{0,Mn} = 14.0 \pm 0.3$ b are used in this study. The procedures used in the determinations of σ_0 and I_0 were described in detail in the previous works (Karadag et al., 2003; Karadag and Yücel, 2005).

Thermal neutron self-shielding (G_{th}) and epithermal neutron self-shielding (G_{epi}) factors for Al_2O_3–3.4%MnO_2 and Al_2O_3–1.05%Sm_2O_3 diluted samples filled in the tubes (height/diameter ≈ 2) have been calculated as G_{th}=0.913 and $G_{epi} = 0.812$ for Sm, and $G_{th} = 0.997$ and $G_{epi} = 0.903$ for Mn, respectively. For the estimation of these factors valid for the diluted samples, the necessary nuclear data (For instance, resonance parameters, absorption, scattering, total microscopic cross-sections, etc.) are taken from JENDL-3.3 and ENDF/B-VI online data libraries. The percentages of dilution for the samples were experimentally determined in order to obtain good counting statistics in the measurements. In the cadmium covered irradiations, cadmium transmission factor for ^{152}Sm ($F_{Cd} = 0.982$), which accounts for the fact that the specific count rate of a cadmium covered isotope of ^{152}Sm is differ from the specific count rate of the bare isotope induced by epithermal neutrons, has taken into account. Since the Westcott correction factors, g (20 °C) are close to unity such as 1.0004 (Ryves and Zieba, 1974) for ^{55}Mn and 1.003 (Chang, 2006) for ^{152}Sm, these nuclides can be considered a good $1/v$-law behaviour nuclides. The correction factors (F_g) for gamma ray attenuations in Al_2O_3–1.05%Sm_2O_3 sample at 103.18 keV emitted from ^{153}Sm and Al_2O_3–3.4%MnO_2 sample at 846.7 keV emitted from ^{56}Mn at a fixed geometry for the case of a cylinder, coaxially positioned with the detector have been calculated to be 1.09 and 1.03, respectively. In the calculations, the simple relation, $F_g = \mu x/(1 - e^{-\mu x})$ is used, where x is sample thickness (cm) and μ is the linear attenuation coefficient (cm^{-1}) of the used mixture compounds determined by using the total mass attenuation coefficients, μ/ρ (cm^2/g) for the mixtures, which are taken from the XCOM database (Berger et al., 2005).

In the RI determination, the effective cadmium cut-off energy is set at 0.55 eV for a detector, having a $\sigma(v) \sim 1/v$ cross section for the (n,γ) reaction up to 1–2 eV, irradiated in an isotropic neutron flux as a small sample in a cylindrical Cd box (height/diameter ≅ 2) with a wall thickness of

Table 2
Experimental uncertainties for the TNX measurements

Uncertainties due to	Uncertainties (%)	
	^{152}Sm	^{55}Mn
Statistical error[a]	0.18	0.39
Detection efficiency	2.55	2.75
Mass of sample	0.01	0.01
Half-life	0.45	0.004
Isotopic abundance	0.60	–
γ-ray Emission probability	1.34	0.30
Thermal neutron self-shielding factor	0.1	0.1
Cadmium transmission factor	0.80	–
Monitor thermal neutron cross section	–	0.75
Total uncertainty	3.09	2.89

[a] Errors are based on counting statistics of ±1.65 σ.

Table 1
Nuclear data used in the experimental analyses

Nuclear reaction	Cd transmission factor[e], F_{Cd}	Effective resonance energy[e], $\bar{E}_r$ (eV)[c]	Half-life (h)[d]	The measured γ-ray[d]	
				Energy (keV)	Emission probability (%)
^{55}Mn(n,γ)^{56}Mn	1.00[a]	468 ± 51	2.5789(1)	846.754(20)	98.9(3)
^{152}Sm(n,γ)^{153}Sm	0.982[b]	8.53 ± 0.09	46.50(21)	103.18(17)	29.8(4)

[a] El Nimr et al. (1981).
[b] El Nimr and Ela-Assaly (1987).
[c] Jovanovic et al. (1987); Kolotov and De Corte (2003); De Corte and Simonits (2003) and Rajput (2003).
[d] NuDat (Sonzogni, 2005).
[e] For $E_{Cd} = 0.55$ eV.

Table 3
Experimental uncertainties for the RI measurements

Uncertainties due to (x_j)	Relative uncertainty, s_j (%)	Error propagation factor, $Z(x_j)$	Relative uncertainty on the RI value, $[s_j \times Z(x_j)$ (%)]
α-shape parameter	19.28	0.13	2.41
Cadmium cut-off energy	15.00	0.04	0.58
Cadmium ratio of ^{56}Mn	1.60	1.07	1.71
Cadmium ratio of ^{153}Sm	1.81	1.79	3.24
Thermal neutron self-shielding factor for Mn sample	0.10	1.00	0.10
Thermal neutron self-shielding factor for Sm sample	0.10	1.00	0.10
Epithermal neutron self-shielding factor for Mn sample	0.20	1.00	0.20
Epithermal neutron self-shielding factor for Sm sample	2.50	1.00	2.50
Reference resonance integral cross-section of ^{55}Mn	2.14	1.46	3.13
Reference thermal neutron cross section of ^{55}Mn	0.75	1.46	1.10
Reference thermal neutron cross section of ^{152}Sm	4.23	1.00	4.23
Effective resonance energy of ^{55}Mn	10.90	0.04	0.45
Effective resonance energy of ^{152}Sm	1.10	0.08	0.09
Total uncertainty, S_T (%)			7.42

Table 4
Thermal neutron cross section (TNX) and resonance integral (RI) values for ^{152}Sm(n,γ)^{153}Sm reaction

Year	References	TNX value, σ_0 (b)	RI value, I_0 (b)	Cadmium cut-off energy, E_{Cd} (eV)
	This work	204.8 ± 7.9	3038 ± 214	0.55
2005	ENDF/B-VI (2005)	205.88	2968.41	0.50
2005	JEFF-3.1 (2005)	205.952	2990.92	
2003	Kolotov and De Corte (2003)	202 ± 7.27	2909 ± 122	0.55
2003	De Corte (2003)	202 ± 7.27	–	0.55
2002	JENDL-3.3 (Shibata et al., 2002)	206.2	2764	
1998	BROND-2.0 (1998)	206	2982.9	0.50
1997	Kafala et al. (1997)	223 ± 10	2962 ± 54	0.55
1989	De Corte and Simonits (1989)	220 ± 5.3	3168 ± 101.4	0.55
1987	IAEA (1987)	206 ± 6	2960 ± 150	
1984	Simonits et al. (1984)	210 ± 11	3020 ± 163	0.55
1984	Mughabghab (1984)	206 ± 6	2970 ± 100	0.50
1978	Heft (1978)	204 ± 9	3050 ± 360	0.50
1976	Federova et al. (1976)	–	2970 ± 80	0.50
1975	Schenter (1975)	–	3007.6	0.50
1974	Van Der Linden et al. (1974)	–	3141 ± 157	0.55
1974	Pope (1974)	–	3462	0.55
1973	Sakata and Nagayama (1973)	–	2700	0.50
1973	Lautenbach (1973)	–	3115	0.465
1972	Steinnes (1972)	210 ± 10	2530 ± 150	0.50
1972	Clayton (1972)	–	3240	0.50
1972	Rahn et al. (1972)	–	2644 ± 604	
1971	BNL (1971)	–	3360	0.50
1969	Rahn and Ho (see Gryntakis and Kim, 1983)	–	2635 ± 598	0.414
1969	Hayodom et al. (1969)	–	2920	0.50
1969	Walker (1969)	–	3000	
1968	Dobrozemsky et al. (1968)	–	2920	
1967	OAEP (1967)	–	2920	
1967	Palmucci (1967)	–	3087	0.50
1962	Garrison and Roos (1962)	–	2230 ± 120	0.40
1962	Cabell (1962)	209 ± 9	3162 ± 104	0.50
1962	Bernabei et al. (1962)	209.1 ± 20.7	–	
1961	Chrien (1961)	–	3182	
1961	Walker and Green (1961)	–	3155	
1960	Tattersall et al. (1960)	224 ± 7	2946 ± 300	0.67
1960	Nephew (1960)	–	2938 ± 300	
1960	Fehr and Hansen (1960)	215 ± 10	2740 ± 150	0.50
1958	Rose et al. (1958)	–	3009 ± 300	
1958	Pattenden (1958)	200 ± 6	–	–
1956	Macklin and Pomerance (1956)	–	1750	0.40
1956	Walker (1956)	250 ± 50	–	–
1950	Harris et al. (1950)	–	1715	0.60

1 mm (Goldstein et al., 1961). Nuclear data used for the radioactivity measurements for TNX and RI determinations are given in Table 1.

For the TNX measurement, the experimental uncertainties for the ^{152}Sm and ^{55}Mn are given in Table 2. The main sources of the uncertainties for the TNX measurement are due to detection efficiencies (2.55% for ^{152}Sm and 2.75% for ^{55}Mn), statistical errors (0.18% for ^{152}Sm and 0.39% for ^{55}Mn), γ-ray emission probability for 103.18 keV gamma peak of ^{152}Sm (1.34%) and cadmium transmission factor (0.80%) and isotopic abundance (0.60%) for ^{152}Sm. The data obtained with different irradiation and counting times of activation samples were relatively close to each other, and the difference of the results is within about 1.3%. A consistency is found among the measured data.

As to the RI measurement, relative uncertainty s_j (%), error propagation factor $Z(x_j)$ and relative uncertainty $[s_j \times Z(x_j)$ (%)] on the present RI value for each parameter and the resulted total uncertainty S_T (%) for the obtained RI value for the reactions of interest are given in Table 3, in which the total relative uncertainty on the obtained RI value has been determined to be $S_T = 7.42\%$ by the error propagation procedure (Karadag and Yücel, 2005).

The TNX value for the ^{152}Sm(n,γ)^{153}Sm reaction has been found to be $\sigma_0 = 204.8 \pm 7.9$ b and given in Table 4, together with other literature values. The present result for the TNX for the ^{152}Sm(n,γ)^{153}Sm reaction, in general, is in good agreement with the measurement of Heft (1978), Kolotov and De Corte (2003), Mughabghab (1984), IAEA (1987) and with evaluated ones in JEFF-3.1 (2005), ENDF/B-VI (2005), JENDL-3.3 (Shibata et al., 2002) and BROND-2.0 (1998). It is close to within 1.5–5% with the values reported by Simonits et al. (1984), Steinnes (1972), Cabell (1962), Bernabei et al. (1962), Fehr and Hansen (1960) and Pattenden (1958), but disagrees with the measurements of Kafala et al. (1997), De Corte and Simonits (1989), Tattersall et al. (1960) and Walker (1956) by 7–22%.

The present RI value for ^{152}Sm(n,γ)^{153}Sm has been found to be $I_0 = 3038 \pm 214$ b, defining the cadmium cut-off energy as 0.55 eV, and is given in Table 4, together with other results appeared in literature. The present RI value for ^{152}Sm(n,γ)^{153}Sm reaction is in good agreement with the values obtained by Simonits et al. (1984) with Cd cut-off energy of 0.55 eV, and the discrepancies between Kolotov and De Corte (2003), Kafala et al. (1997), De Corte and Simonits (1989), Van Der Linden et al. (1974) with assumed Cd cut-off energy of 0.55 eV, and the present measurement are within ≈2.5–4.3%. However, with Cd cut-off energy of 0.55 eV, the present result disagrees with the value reported by Pope (1974) by 14%. The discrepancies between the present RI value and the earlier experimental data for ^{152}Sm(n,γ)^{153}Sm reaction with assumed Cd cut-off energy of 0.50 eV are about 0.4–9.5%. However the present result disagrees within 26–43% with the measurement of Garrison and Roos (1962), Macklin and Pomerance (1956) and Harris et al. (1950).

4. Conclusion

The obtained result for the TNX value of the ^{152}Sm(n,γ)^{153}Sm reaction, generally, is in good agreement, within the limits of error, with the most values appeared in literature. As well, the present RI value at the definition of Cd cut-off energy, 0.55 eV for this reaction obtained by this experiment agrees well with the newest several experimental values, 3020 ± 163 b by Simonits et al. (1984), 3141 ± 157 by Van Der Linden et al. (1974) and 2962 ± 54 b by Kafala et al. (1997), Kolotov and De Corte (2003), within about 2.5–4.5%. However, the present RI value disagrees with some earlier measurements by 43%.

It seems that since the RI value of the ^{152}Sm(n,γ)^{153}Sm reaction are relatively high, a radioactive neutron source such as ^{241}Am–Be having with relatively low neutron flux can be chosen a convenient alternative for the activation method with a different monitor such as ^{56}Mn isotope besides standard ^{198}Au monitor with favourable other nuclear properties. In spite of well-known good nuclear properties of standard ^{198}Au monitor, the ^{56}Mn monitor isotope has been used for the RI determination in this work, considering the fact that the well separated principal resonance energy of ^{197}Au monitor lies in 4.9 eV, which is very close to $1/v$ thermal energy region, and whereas the first resonance of ^{55}Mn lies in 337 eV, which is quite far from $1/v$ region. In conclusion, the reasonable consistency between the newest several experimental values other monitors and the present result using Mn-monitor in non-ideal epithermal neutron spectrum for the RI value for the ^{152}Sm(n,γ)^{153}Sm reaction implies that Mn-monitor may also be considered as an alternative comparator for the especially RI determinations.

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