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Determination of effective resonance energies for the (n,γ) reactions of ^{152}Sm and ^{165}Ho by using dual monitors

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ABSTRACT

The effective resonance energies $\overline{E}_r$ for the (n,γ) reactions of ^{152}Sm and ^{165}Ho isotopes were determined by using dual monitors (^{55}Mn–^{98}Mo) due to their favourable resonance properties. The samples were irradiated in an isotropic neutron field obtained from ^{241}Am–Be neutron sources. The induced activities were measured with a high efficient, p-type Ge detector. The necessary correction factors for thermal neutron self-shielding (G_{th}), resonance neutron self-shielding (G_{epi}), self absorption (F_s) and true coincidence summing (F_{coi}) effects for the measured γ-rays were taken into account. Thus, the experimental $\overline{E}_r$-values for above (n,γ) reactions are found to be 8.65 ± 1.80 eV for ^{152}Sm and 12.90 ± 2.69 eV for ^{165}Ho isotopes, respectively. The $\overline{E}_r$-values for both ^{152}Sm and ^{165}Ho isotopes were also theoretically calculated from the newest resonance data in the literature. Theoretically calculated $\overline{E}_r$-values are estimated to be 8.34 eV and 8.53 eV for ^{152}Sm by two different approaches, which are generally, much smaller than that the present experimental value by 1.4–3.6% for ^{152}Sm. In case of ^{165}Ho isotope, the theoretically calculated $\overline{E}_r$-value of 8.63 eV from the first approach deviates substantially from the measured value by about 33%, whereas the theoretical $\overline{E}_r$-value of 12.95 eV from the second approach agrees very well with our experimentally determined $\overline{E}_r$-value. The results show that the present experimental $\overline{E}_r$-values for ^{152}Sm and ^{165}Ho isotopes agree with the calculated ones from the second approach within limits of the estimated uncertainty if the recently evaluated resonance data are used. However, it is worth noting that the results for $\overline{E}_r$-value calculated from the first approach are not satisfactorily accurate because of neglecting the neutron widths in that approach. Therefore, this study implies that it be regarded to the experimentally determined $\overline{E}_r$-value introduced in k_0–NAA method for the determination of any analytical result rather than its theoretical value.

1. Introduction

In recent years, Holmium (Ho) and Samarium (Sm) are commonly used rare-earth elements that are attractive for therapeutic radionuclide production. For instance, ^{166m}Ho (1.21×10^3 years) and ^{166g}Ho (26.83 h) could be produced from the neutron capture reaction of ^{165}Ho (100%) stable isotope. The first one is the long-lived isotope which is generally used in laboratory calibration source. But the latter one is mostly used in radiotherapy due to its good decay properties such as high beta energies 1.773 MeV (48.7%) and 1.854 MeV (50.0%) and its prominent γ-ray energy 80.574 keV (6.71%) [1,2]. It is reported that ^{166g}Ho due to favourable decay characteristics could be used in endovascular radionuclide therapy technique in liquid filled low pressure balloon angioplasty, which is a well known standard treatment for atherosclerotic coronary artery disease [3]. On the other hand, Sm is also mostly used in nuclear reactors as an absorber material because of high thermal and epithermal neutron cross sections. Apart from this, some Samarium isotopes have also a great importance in nuclear medicine for therapeutic purposes. For example, the radioactive isotope ^{153}Sm, which is produced from ^{152}Sm(n,γ) reaction, is used as one of the β^- emitting therapeutic radioisotope in nuclear medicine for tumour therapy and bone pain palliation due to its high local beta dose per disintegration and suitable half-life (46.284 h) with beta energies 0.635 MeV (32.2%) and 0.705 MeV (49.6%), Because ^{153}Sm disintegrates beta transitions to excited levels and to the ground state of ^{153}Eu by emission of the prominent γ-rays with 69.67 keV (4.85%) and 103.18 keV (30%) energy, while other γ-ray emissions with energy up to 763.8 keV are relatively weak [4]. Additionally, it is reported that ^{153}Sm has been tested and used in insect ecology (insects

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behaviors, 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 [5].

The motivation for the present measurements for the reactions ^{165}Ho(n,γ)^{166g}Ho and ^{152}Sm(n,γ)^{153g}Sm was the discrepancies among effective resonance energies. Because the effective resonance energies as well as thermal neutron and resonance integral cross sections are important in NAA analysis, e.g. in k_0–NAA method. Additionally neutron activation cross section data are also commonly used in other studies related to the interaction of neutrons with matter [6–8]. On the other hand, since the launching of the NAA k_0-standardization concept [9], great efforts have been made to perform NAA technique in absolute or single-comparator standardization mode for the neutron capture (n,γ) activation analysis of any analyte. However, single comparator and absolute standardizations suffer from two important drawbacks [10]. The first one is that the inflexibility of single comparator method is strictly bound to a given set of local irradiation and counting conditions. The latter one, i.e., absolute standardization method is caused by the introduction of occasionally unreliable absolute nuclear data for activation and decay. In the neutron capture (n,γ) activation analysis, for instance, the absolute standardization (e.g. the k_0–NAA method) uses the concept of effective energy $\overline{E}_r$ to correct for the effect of non-ideality of the epithermal neutron flux distribution, which is often represented by a non-ideal $1/E^{1+\alpha}$ with a spectrum shape factor, α. It is well known that in case non-ideality of epithermal spectrum shape (deviation from ideal $1/E$ behaviour) in an irradiation position has been neglected, this may lead to significant errors on the analytical result due to the inaccuracy of the essential nuclear parameter $I_0(\alpha)$, which is resonance integral cross section [11]. As well as the epithermal spectrum shape factor (α), the effective resonance energy ($\overline{E}_r$) parameter of the nuclide to be activated is also another essential parameter for the correction of resonance integral to thermal neutron cross section ratio, $Q_0(\alpha)$ ($=I_0(\alpha)/\sigma_0$) in a real $1/E^{1+\alpha}$ epithermal flux distribution [12,13]. Therefore, the inaccurate of effective resonance energy ($\overline{E}_r$)value can also give rise to more serious error on the analytical result because it is directly related to the $1/(\overline{E}_r^{\alpha})$ in the $I_0(\alpha)$ definition [11,14].

From point of view of accurate NAA analysis, the question arises whether the used $\overline{E}_r$-values used are susceptible to updates of experimental resonance data due to their dependencies on individual resonance energies and resonance widths. The extent of the experimental instability of $\overline{E}_r$-values, as well as the degree of inaccuracy of the resonance integral values need to look into carefully the experimental determination of $\overline{E}_r$-values [15]. In addition, it is reported that, in some cases when the theoretical resonance data for $\overline{E}_r$-values are still incomplete, obsolete, inaccurate or even not known at all. Hence, the experimental determination of the $\overline{E}_r$-value for an isotope should be carried out [11].

It is therefore that in the present study it is aimed to determine the new experimental $\overline{E}_r$-values for the particular (n,γ) reactions of ^{165}Ho and ^{152}Sm target isotopes by using two different monitors (^{55}Mn–^{98}Mo). The determination of$\overline{E}_r$-values is based on the well-reliable cadmium ratio (Cd-ratio) measurements of the isotope being investigated since the Cd-ratio method greatly allows to obtain the reproducible results and thus eliminate the possible differences in the neutron spectrum between the thermal and epithermal activation on the condition that same irradiation and counting conditions is applied to each measurement.

On the other hand, since the experimentally determined $\overline{E}_r$-values for ^{165}Ho and ^{152}Sm isotopes do not appear at all in surveying literature, those $\overline{E}_r$-values theoretically calculated for the above isotopes using the recent resonance parameter data could also be compared with each other. It is thought that this would be helpful in determination of $\overline{E}_r$-values since the quantity and quality of the available resonance parameters' data in the present literature improve continuously.

2. Experimental

Each of the samples was separately mixed with a sufficient amount of Al_2O_3 powder to reduce to some degree the neutron self shielding effects. Then, the samples (Al_2O_3 + Ho_2O_3 and Al_2O_3 + Sm_2O_3) were filled in the 1 mm thick polystyrene tubes (6.5 mm radius and 6.25 mm height). They were exposed to the neutrons in a fixed position of the irradiation hole. The samples were irradiated with 1 mm thick cylindrical Cd filter to obtain Cd-ratio values. The percentages of dilution for the samples were experimentally determined in order to obtain optimum counting statistics in the measurements. Ten samples for each element were individually prepared as given in Table 1, together with the percentage dilution, and their neutron self-shielding factors. The thermal and epithermal self-shielding factors (G_{th} and G_{epi}) for the powder mixtures filled in the polystyrene sample tubes were calculated using the procedures for the case of the irradiations in an isotopic neutron field [6,16–18]. But those factors for the case of Au and Mo-foils exposed to the same neutron field were calculated by the Nisle' approximation [19]. The required nuclear data (e.g. resonance parameters, absorption, scattering, total microscopic cross-sections, etc.) were taken from JENDL-3.3 [20] and NUDAT [21] online data libraries.

A set of five samples was prepared for each element for obtaining Cd covered-irradiation data. The remaining a set of five samples for each element was then used for obtaining bare irradiation data. In addition, the thin foils of Au and Mo were also irradiated as monitor isotopes.

The irradiations were performed in an isotropic neutron field obtained from ^{241}Am–Be neutron sources with total activity 3×592 GBq. The sources were immersed in a paraffin moderator and shielded with 1 mm thick Cd sheet and lead bricks, which was installed at ex-Ankara Nuclear Research and Training Center (ANRTC). The detailed geometrical configuration of this neutron irradiator has been previously described, elsewhere [7,16,22].

The irradiated samples were measured by using a γ-ray spectrometer with a p-type, coaxial high pure Ge detector (Canberra Model GC 11021) 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 Ge crystal has 82 mm diameter and 85.5 mm length mounted in a 101.6 mm diameter Al-end cap. The detector was installed in a 10 cm thick Pb shield lined with a 1 mm thick Sn and 1.5 mm Cu thick (Canberra Model 767) for reducing Pb X-rays in the 72–88 keV. The lead shield was also jacketed by a 9.5 mm steel outer housing. The detector was interfaced to a digital spectrum analyzer (Canberra DSA-1000) with a full featured 16 K ADC/MCA analyzer, based on digital signal pro-

Table 1
The calculated neutron self-shielding factors for the used samples.

Irradiated samples	Target isotope	Thermal neutron self shielding factor including scattering, G_{th}	Epithermal neutron self shielding factor, G_{epi}
Al_2O_3-3.4% MnO_2	^{55}Mn	0.997	0.903
Al_2O_3-1.05% Sm_2O_3	^{152}Sm	0.913	0.812
Al_2O_3-1.2% Ho_2O_3	^{165}Ho	0.997	0.890
Mo 0.025 mm-foil	^{98}Mo	0.999[a]	0.992[b]
Au 0.0005 mm-foil	^{197}Au	1.000[a]	0.925[c]

[a] Calculated by Nisle approximation, including scattering effect, given in Gilat and Gurfinkel (1963) [19].
[b] Calculated by approximation given in Beckurtz and Wirtz (1964) [23].
[c] IAEA (1970) [24].