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Determination of effective resonance energies for the ^{185}Re(n,γ)^{186}Re and ^{187}Re(n,γ)^{188}Re reactions by cadmium ratio method

Mustafa Güray BUDAK*
Department of Mathematics and Science Education, Gazi Faculty of Education, Gazi University, Ankara, Turkey

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Abstract: The effective resonance energy ($\bar{E}_r$) values for the ^{185}Re(n,γ)^{186}Re and ^{187}Re(n,γ)^{188}Re reactions were measured by using the cadmium ratio method. ^{197}Au and ^{98}Mo isotopes were selected as dual monitors to determine the epithermal neutron spectrum shape factor in the irradiation sites of a 37 GBq ^{241}Am-Be neutron source. The analytical grade rhenium(VII) oxide samples were diluted with Al_2O_3 powder to reduce neutron self-shielding effects, and they were put into 1-mm thick small cylindrical polytetrafluoroethylene (PTFE) boxes. Samples were irradiated in a ^{241}Am-Be neutron source. The activation produced in the irradiated samples was measured in close-counting geometry on the end cap of a Ge detector in order to obtain better counting statistics. The correction factors for gamma-ray self-absorption (F_s), thermal neutron self-shielding (G_{th}), and epithermal neutron self-shielding (G_{epi}) effects were determined with suitable approaches. Consequently, the experimental $\bar{E}_r$-values were measured to be 3.49 ± 0.54 eV for ^{185}Re and 41.77 ± 6.79 eV for ^{187}Re target nuclide, respectively. Furthermore, the $\bar{E}_r$-values were theoretically computed through up-to-date resonance data obtained from the ENDF/B VII library using two different approaches. Since there is no experimental data available in the literature for the $\bar{E}_r$-values of these isotopes, the results were compared with the theoretical values in the literature.

Key words: Effective resonance energy, neutron activation, cadmium ratio, epithermal spectrum shape factor, rhenium

1. Introduction

In recent years, k_0-NAA has become a well-established neutron activation analysis (NAA) method as a nuclear analytical technique [1]. However, the k_0-NAA method needs nuclear data such as effective resonance energy $(\bar{E}_r)$ and Q_0 $(=I_0/\sigma_0)$. $\bar{E}_r$ represents "the energy of a single virtual resonance which gives the same resonance activation rate as all actual resonances for the isotope" [2]. The term Q_0 is the ratio of the total resonance integral to the 2200 m/s cross-section (I_0/σ_0).

The literature review reveals that the first calculations for $\bar{E}_r$ values date back to 1979 and 1987. As a measurement technique, a multichannel method was suggested for simultaneous experimental determination of Q_0 and $\bar{E}_r$ in 1984 [3]. No new measurements have been made for $\bar{E}_r$ values of some isotopes for about 30 years [4]. In addition, there are no $\bar{E}_r$ values for some isotopes in the literature available today.

In this study, the $\bar{E}_r$-values for the ^{185}Re(n,γ)^{186}Re and ^{187}Re(n,γ)^{188}Re reactions were measured using the cadmium ratio method. ^{185}Re (37.4%) and ^{187}Re (62.6%) isotopes have good nuclear properties in terms

*Correspondence: mgbudak@gmail.com

of neutron activation analysis. The epithermal spectrum shape factor of a neutron field is a key parameter in characterizing the neutron field in the sample irradiation position. ^{197}Au and ^{98}Mo monitor isotopes were selected for the determination of α shape factor. The Cd ratio method has been chosen because it removes the effects of variations in the spectrum on our measurements.

2. Materials and methods

2.1. Theoretical calculation of $\bar{E}_r$-value

The effective resonance energy for an isotope can be calculated according to the Breit–Wigner formula [5], which considers all resonances depending on resonance neutron energies.

$$\left(\bar{E}_r\right)^{-\alpha} = \frac{\sum\limits_i \frac{\pi}{2} \frac{\sigma_i \ \gamma,i \ E_{r,i}^{-\alpha}}{E_{r,i}}}{\sum\limits_i \frac{\pi}{2} \frac{\sigma_i \ \gamma,i}{E_{r,i}}}. \tag{1}$$

$\bar{E}_r$ is not a real nuclear constant for an isotope since it is a function of α. However, when $\left(\bar{E}_r\right)^{-\alpha}$ is extended in a Taylor series, the higher-order terms of the series can be neglected because of their small values. This approximation was suggested for the first time by Moens et al. [5] to estimate the effective resonance energy of an isotope, as in the following equation:

$$\ln \bar{E}_r = \frac{\sum\limits_i \frac{\sigma_i \ \gamma,i \ln E_{r,i}}{E_{r,i}}}{\sum\limits_i \frac{\sigma_i \ \gamma,i}{E_{r,i}}}, \tag{2}$$

where $\bar{E}_{ri}$ is the i-th resonance energy, σ_i is neutron capture cross-section at the $\bar{E}_{ri}$ energy, and $\Gamma_{\gamma,i}$ is the radiative width of the resonance energy. Eq. (2) assumes that resonance self-shielding effects are eliminated when sufficiently diluted samples are used. The current data needed for calculations are taken from the ENDF/B VII database [6]. Figures 1 and 2 exhibit the resonance cross-section data of ^{185}Re and ^{187}Re. The main disadvantage of this approach is that neutron widths (Γ_n) are not taken into account in the equation.

The second approach for the theoretical calculation of $\bar{E}_r$-value was proposed by Jovanovic et al. [7]:

$$\ln \bar{E}_r = \frac{\sum\limits_i w_i \cdot \ln E_{r,i}}{\sum\limits_i w_i}. \tag{3}$$

In Eq. (3), the symbol of w_i stands for the weighting factors.

$$w_i = \left(\frac{g \ \gamma \ n}{}\right)_i \cdot \frac{1}{E_{r,i}^2}. \tag{4}$$

In this equation, g = (2j + 1) / 2(2I + 1) is the statistical weight factor, Γ_n is the neutron width, Γ_γ is the radiative width, Γ is the total width of resonance which is the sum of Γ_n and Γ_γ, and J and I are the spin of the resonance state of the neutron captured compound nucleus and the target nucleus. The resonance data used in the calculations were taken from current literature [6].

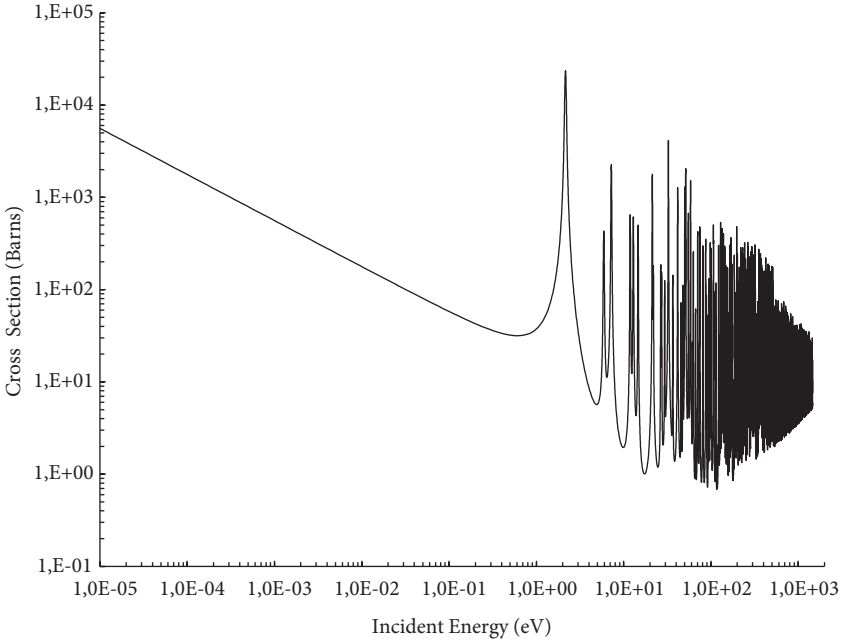

Figure 1. Cross-section versus incident neutron energy for ^{185}Re [6].

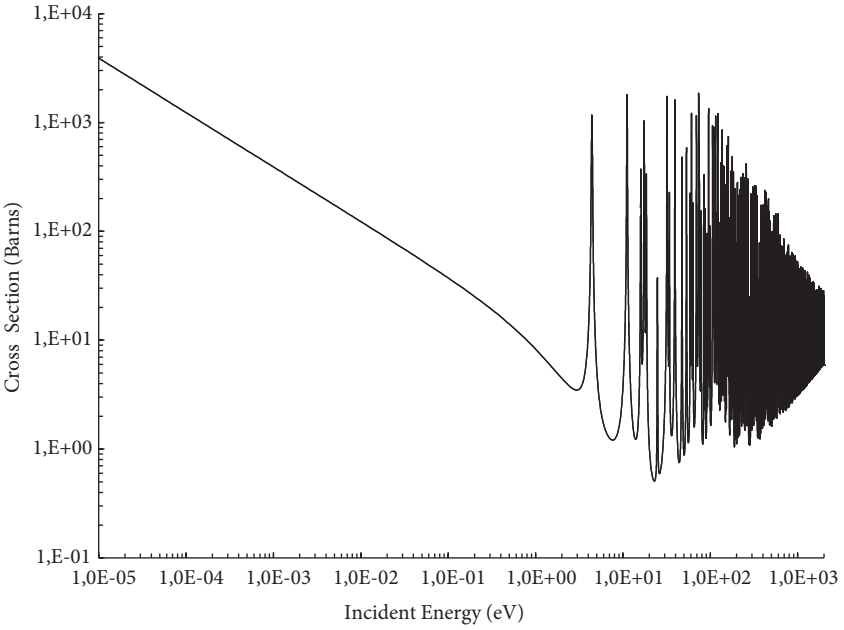

Figure 2. Cross-section versus incident neutron energy for ^{187}Re [6].

2.2. Experimental method

The detailed description of the currently used neutron irradiation unit has already been given in Yücel et al. [8]. The unit consisting of a 37 GBq ^{241}Am-Be source was placed in the center of a tank filled with water. The neutron source was then moderated using a chamber filled with paraffin. This chamber was also coated with paraffin blocks loaded with boron oxide. This entire structure was plated with a boron oxide layer to prevent

neutron leakage. A schema of the experimental system used in this study is shown in Figure 3. The neutron source was in a fixed position in the middle of the two irradiation cells. One of the irradiation cells is called Site #1; it was equipped with a pneumatic transfer system. The other cell that was manually controlled is Site #2. In Site #1, the sample was transferred to the irradiation position by an air compressor, and was taken back to the laboratory under suitable air pressure.

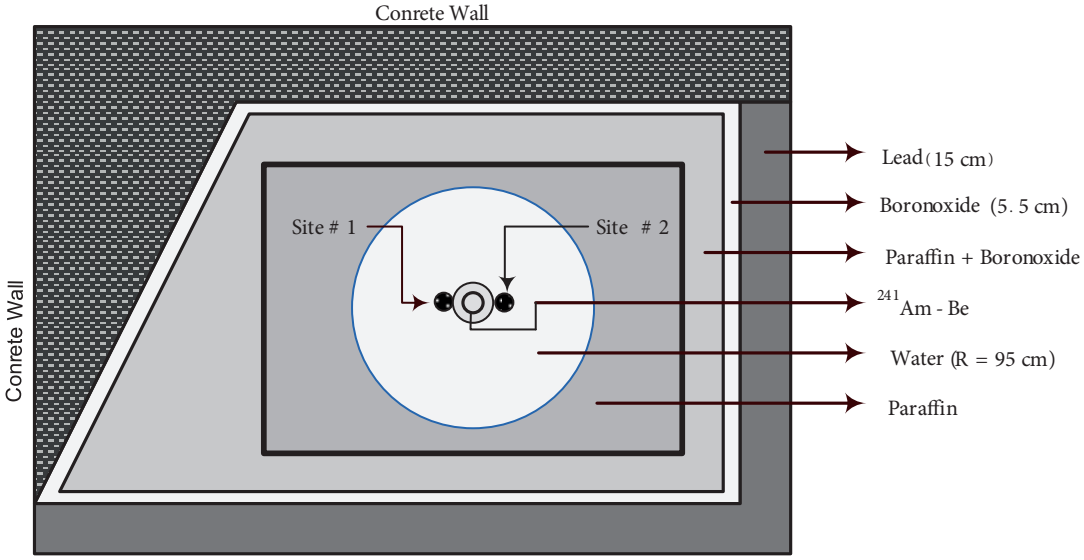

Figure 3. A schematic view of the neutron irradiation unit with a 37 GBq ^{241}Am-Be source [8].

Au and Mo foils and samples of diluted Re_2O_7 were irradiated in both sites. The experimental process was performed using two sets of samples. One set was composed of bare samples, that is, polytetrafluoroethylene (PTFE) boxes were sent to one irradiation site. The other set of samples was irradiated within 1-mm thick Cd cylinder boxes.

The neutron spectrum characterization parameters of the irradiation cells were determined through experiments in a previous study [8]. The epithermal spectrum shape factor was $\alpha = 0.045 \pm 0.009$ for the irradiation position at Site #1, and it was $\alpha = 0.038 \pm 0.008$ at Site #2. The thermal to epithermal flux ratio was f $= 63.6 \pm 1.5$ for the irradiation position at Site #1, and it was f $= 50.9 \pm 1.3$ at Site #2.

The specific activities of ^{99}Mo were derived in two different cases: in the first case foils were irradiated bare, whereas in the second case they were irradiated within the 1-mm thick Cd sheaths. According to the decay scheme of the product (^{99}Mo $[\beta^-]$ ^{99m}Tc) after neutron capture, the ^{99m}Tc radionuclide emits the most intense gamma rays at 140.511 keV with the emission rate of 88.5% ± 2%. For this reason, this gamma ray has been specially selected to derive the specific activity of ^{99}Mo. In addition, the ^{99}Mo isotope contributes to the 140.511 keV peak area (the emission probability is 4.52 ± 0.24%). Therefore, it is necessary to consider branching ratio correction to achieve more accurate results [9]. After this correction, the net specific activity of ^{99}Mo was calculated using the process recommended by Simonits et al. [10,11].

In this study, a complete multichannel analyzer consisting of a p-type Ge well-type detector with a 44.8% relative efficiency (Canberra, GCW4023) and its associated electronics were used to acquire the gamma-ray spectra.

The k_0 method considers the notion of $\bar{E}_r$ in order to fix the effect of the nonideal structure of the

Table 1. Nuclear data for the isotopes.

Target isotope (abundance)	Nuclear reaction	Cadmium transmission factor, F_{Cd}[a]	Effective resonance energy, E_r (eV)[a]	Q_0 (=I_0/σ_0)[a,b] S (%)	Half-life[b]	The measured gamma-ray[c,d]		Self-attenuation factor	Thermal neutron self-shielding factor[f] G_{th}	Epithermal neutron self-shielding factor[f] G_{epi}
						Energy (keV)	Emission probability, γ (%)	F_s (From NIST)[e]		
^{197}Au (100%)	^{197}Au(n,γ)^{198}Au	0.992	5.65 ± 0.40	15.7 (1.8%)	2.272 (16) d	411.802 (17)	95.62 (6)	1.0005	0.999	0.922
^{98}Mo (24.13%)	^{98}Mo(n,γ)^{99}Mo ^{98}Mo(n,γ)^{99}Mo(β)^{99}Tc	1.000	241 ± 48	53.1 (6.3%)	65.976 (24) h 6.0067 (5) h	140.511 (1) 140.511 (1)	4.52 (24) 88.5 (2)	1.0011 1.0011	1.000	0.945
^{185}Re (37.4%)	^{185}Re(n,γ)^{186}Re	1.000	-	15.4 (2.5%)	3.7183 (11) d	137.157 (8)	9.42 (6)	1.0143	0.999	0.979
^{187}Re (62.6%)	^{187}Re(n,γ)^{188}Re	1.000	-	4.34 (6.4%)	17.004 (22) h	155.041 (4)	15.2 (6)	1.0123	0.999	0.979

[a]De Corte and Simonits [19].

[b]Kolotov and De Corte [20].

[c]NUDAT 2.7 [21].

[d]NUCLÉIDE LARA database [22].

[e]Self-attenuation calculated using mass attenuation coefficients taken from NIST [23].

[f]Calculated by approximations given in Chilian et al. [17] and Salgado et al. [18].

epithermal flux distribution. $\bar{E}_r$ is defined as [3,5]:

$$\bar{E}_{r,i} = \left[(Q_0 - 0.429) \Big/ \left(\frac{f}{F_{Cd} \cdot R_{Cd} - 1} - \frac{0.429}{(2\alpha + 1) \cdot E_{Cd}^{\alpha}} \right) \right]^{1/\alpha} \tag{5}$$

where $\alpha$is the shape factor that stands for the deviation from ideal 1/E spectrum and f is the thermal-to-epithermal flux ratio. When the epithermal spectrum of the neutron source is nonideal, neglecting deviation from the 1/E behavior can lead to significant errors in analytical results [3].

$\bar{E}_r$ of the isotopes of interest can be determined using the following equation where c represents the parameters of the comparator [3,12].

$$\bar{E}_r = \bar{E}_{r,c} \left[\frac{Q_0 - 0.429}{Q_{0,c} - 0.429} \cdot \frac{\frac{f}{(F_{Cd,c} \cdot R_{Cd,c} - 1) \cdot \left(\frac{G_{epi}}{G_{th}} \right)_c} - C_\alpha}{\frac{f}{(F_{Cd} \cdot R_{Cd} - 1) \cdot \left(\frac{G_{epi}}{G_{th}} \right)} - C_\alpha} \right]^{1/\alpha} \tag{6}$$

where $C_\alpha = \frac{0.429}{(2\alpha+1) \cdot E_{Cd}^{\alpha}}$ and $0.429 \approx 2 \cdot \sqrt{E}_0 / E_{Cd} = 2 \cdot \sqrt{0.025}/0.55$.

As noted in the earlier studies [13–16], $R_{Cd} = A_{sp}^{-} \big/ A_{sp}^{+}$ ratio can be calculated using the specific activities measured in experiments. The self-shielding factors (thermal, G_{th} and epithermal, G_{epi}) used in Eq. (6) were computed using approaches in the studies of Chilian et al. [17] and Salgado et al. [18]. Necessary nuclear data was obtained from the ENDF/B VII.1 online database [6]. Decay and nuclear data used in the determinations are listed in Table 1.

3. Results

At the end of this experiment, the new experimental $\bar{E}_r$-values for the ^{185}Re(n,γ)^{186}Re and ^{187}Re(n,γ)^{188}Re reactions were determined using the reliable cadmium ratio method. The results obtained for $\bar{E}_r$ values are listed in Table 2, where it is shown that there is a consistency between the results obtained when ^{197}Au and ^{98}Mo dual monitors are used. The weighted $\bar{E}_r$ values are given in Table 3 with up-to-date and previous values derived from two different theoretical calculation models. The calculation procedure used to determine the uncertainty of $\bar{E}_r$ was explained in detail in previous studies [13,14,16]. Considering the uncertainties of the parameters in Eq. (6), it can be concluded that it is not possible to reduce the final uncertainty below 20% for the results obtained for $\bar{E}_r$-values.

Table 2. The measured $\bar{E}_r$ for ^{185}Re and ^{187}Re.

Reaction	$\bar{E}_r$(eV) (Experimental)		
	Using ^{197}Au monitor	Using ^{98}Mo monitor	Weighted average
^{185}Re(n,γ)^{186}Re	3.54 ± 0.69	3.39 ± 0.89	3.49 ± 0.54
^{187}Re(n,γ)^{188}Re	42.40 ± 8.58	40.63 ± 11.11	41.77 ± 6.79

In general, the experimentally determined $\bar{E}_r$-values for ^{185}Re and ^{187}Re isotopes are consistent with the values calculated in theoretical approaches. On the other hand, as explained in detail in previous studies, the theoretical approach of Moens [5] (Eq. (2)) does not give accurate and satisfactory results, even current

Table 3. $\bar{E}_r$-values for ^{185}Re and ^{187}Re isotopes.

Reaction	Experimental $\bar{E}_r$ value	Theoretically calculated $\bar{E}_r$ values[a]			
	This work	This work		Literature	
	$\bar{E}_r$ (eV)	$\bar{E}_r$ (eV) Breit–Wigner[b]	$\bar{E}_r$ (eV) Moens[c]	$\bar{E}_r$ (eV) Breit–Wigner[7]	$\bar{E}_r$ (eV) Moens[5]
^{185}Re(n,γ)^{186}Re	3.49 ± 0.54	3.53	2.61	3.40	2.54
^{187}Re(n,γ) ^{188}Re	41.77 ± 6.79	42.20	20.25	41.1	12.0

[a] Since uncertainties are not specified in the data used, the uncertainty of the calculated values is not given.

[b] Calculated by Eq. (3).

[c] Calculated by Eq. (2).

resonance data is used [13,14,16]. The results that are found through this approach are neither correct nor satisfactory, as the approach does not consider the neutron widths. However, the approach suggested by Jovanovic et al. [7] (Eq. (3)) considers the neutron widths, and thus matches the experimental results better.

4. Discussion

In the light of the present findings for ^{185}Re and ^{187}Re, it can be concluded that, especially for isotopes analyzed with the k_0-NAA standardization method, new experimental $\bar{E}_r$-values should be redetermined. It can be said that the cadmium ratio method is a simple but reliable approach to achieve this aim.

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