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MODIFICATION AND GENERALIZATION OF SOME METHODS TO IMPROVE THE ACCURACY OF α-DETERMINATION IN THE $1/E^{1+\alpha}$ EPITHERMAL NEUTRON SPECTRUM

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Some methods described in the literature for the determination of α in the $1/E^{1+\alpha}$ epithermal neutron spectrum are critically reviewed with respect to their accuracy. The multi resonance – detector method with Cd-covered irradiations, as used by SCHUMANN and ALBERT, is generalized by subtracting the epithermal 1/v-tail and by introducing the effective resonance energy, as defined by RYVES. The two-detector method of RYVES is modified by using Cd-ratio measurements, thus eliminating the introduction of systematic errors due to the inaccuracy of absolute nuclear data. The adapted methods are applied in channel 15 of the Thetis reactor (Gent).

Introduction

When dealing with resonance integrals in (n, γ) activation analysis, a problem may arise concerning the effect of the epithermal neutron distribution in the irradiation position under consideration. Although this effect is often overlooked or neglected, it can in some cases have a considerable influence on the value of the resonance integrals for use in a particular irradiation site and it should be considered when resonance integrals are experimentally determined.

If the ideal epithermal neutron spectrum is represented by

$$\Phi_e(E) = \Phi_e \frac{1}{E} \tag{1}$$

where $\Phi_e(E)$ – epithermal flux per unit of neutron energy interval;
Φ_e – energy-independent proportionality constant
= integrated epithermal flux per unit of ln E-interval,

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a resonance integral for this 1/E-epithermal spectrum is written as:

$$I_0 = \int_{E_{Cd}}^{\infty} \frac{\sigma(E)}{E} \, dE \qquad (2)$$

with E_{Cd} – effective Cd cut-off energy
= 0.55 eV for a detector, having a $\sigma(v) \sim 1/v$ activation cross-section for an (n, γ) reaction up to 1–2 eV, irradiated in an isotropic neutron flux as a small sample in a cylindtrical Cd-box (height/dia. = 2) with 1 mm wall-thickness.[1]

The resonance integrals, defined according to Eq. (2) and tabulated in literature, are not valid in a non-ideal, real epithermal neutron spectrum, which can be approximated by:[2,3]

$$\Phi_e(E) = \Phi_e \frac{1 \text{ eV}^{\alpha}}{E^{1+\alpha}} \qquad (3)$$

with α – energy-independent correction factor, which can be positive or negative, depending on the reactor configuration (moderator material, geometry of irradiation site, etc.)

Φ_e – energy-independent proportionality constant; it is now the integrated epithermal flux per unit of $(E^{-\alpha}/-\alpha) \cdot 1 \text{ eV}^{\alpha}$-interval (the fact that this definition is different from that in Eq. (1) has no consequence in the following considerations.)

Accordingly, a resonance integral for a $1/E^{1+\alpha}$ epithermal spectrum should be defined as:

$$I_0(\alpha) = \int_{E_{Cd}}^{\infty} \frac{\sigma(E) \cdot 1 \text{ eV}^{\alpha}}{E^{1+\alpha}} \, dE \qquad (4)$$

Eq. (4) indicates that the resonance integrals for practical use are a function of α, and thus of the irradiation site under consideration. This implies that for conversion of I_0 to $I_0(\alpha)$ and vice versa, one should know the relationship between I_0 and $I_0(\alpha)$, whereas α should be known or experimentally determined.

Using the concept of the effective resonance energy $\bar{E}_r$,[3,4] it can be shown that, for detectors with $\sigma(v) \sim 1/v$ up to 1–2 eV, the following relation holds:

$$I_0(\alpha) = 1\ \text{eV}^{\alpha} \left[\frac{I_0 - 0.426\,\sigma_0}{(\bar{E}_r)^{\alpha}} + \frac{0.426\,\sigma_0}{(2\alpha + 1)\,(E_{Cd})^{\alpha}} \right] \tag{5}$$

where: $\sigma_0 = 2200\ \text{m} \cdot \text{s}^{-1}$ (n, γ) cross-section.

Note that Eq. (5) is only valid for $E_{Cd} = 0.55$ eV, since $0.426 = 2\,(E_0/E_{Cd})^{1/2}$, with $E_0 = 0.025$ eV and $E_{Cd} = 0.55$ eV.

In order to demonstrate the important effect of α on the resonance integrals for some (n, γ) reactions, Table 1 shows $I_0(\alpha)$ for $\alpha = 0.1$, a value which may occur at relatively large distances from the core in a small, graphite-moderated reactor.[3] As compared to I_0 ($\alpha = 0$), differences are observed as high as a factor 1.18 for ^{197}Au, 1.79 for ^{96}Zr and 2.16 for ^{94}Zr.

For the experimental α-determination, various simple methods, based on Cd-covered irradiations, are described in the literature, e.g. the multi resonance – detector method, as used by SCHUMANN and ALBERT,[2] and the two-detector method of RYVES.[3] Since these methods were selected to determine α in the irradiation channels of the Thetis reactor (Gent), for the correction of I_0/σ_0-values in the determination of k_0-factors,[6,7] they were critically reviewed with respect to their accuracy. The present paper deals with the results of this examination, leading to some modifications or generalizations of the described techniques as an attempt to eliminate systematic errors.

Table 1
Effect of a deviating epithermal neutron spectrum ($\alpha = 0.1$) on the resonance integral for some (n, γ) reactions ($E_{Cd} = 0.55$ eV)

Reaction	Effective resonance energy $\bar{E}_r$[4], eV	σ_0, barn	1/E-spectrum I_0 ($\alpha = 0$), barn	$1/E^{1+\alpha}$-spectrum I_0 ($\alpha = 0.1$); Eq.[5], barn
^{197}Au(n, γ)^{198}Au	5.47	98.8	1550	1314
^{96}Zr(n, γ)^{97}Zr	340	0.020	5.0	2.8
^{94}Zr(n, γ)^{95}Zr	4520	0.052	0.300	0.139

Generalization of the multi resonance – detector method, as used by SCHUMANN and ALBERT[2]

In the multi resonance – detector method described by SCHUMANN and ALBERT,[2] use is made of Eq. (3) which can be written as:

$$\Phi_e(E)\,E = \Phi_e\,E^{-\alpha}\;1\;eV^{\alpha} \tag{6}$$

Thus, when plotting log $\Phi_e(E)$ E versus log E, a straight line will be obtained with slope $-\alpha$.

Discrete values for $\Phi_e(E)$ E can be obtained by irradiating, under Cd-cover, a series of resonance detectors, each of them characterized by one dominant resonance peak at energy E_r, with a negligible underlying 1/v-tail. For each detector holds:

$$\Phi_e(E_r)\,E_r = \Phi_e\,E_r^{-\alpha}\;1\;eV^{\alpha} \tag{7}$$

$$= \frac{\Phi_e\,I_0(\alpha)}{I_0(\alpha)}\,E_r^{-\alpha}\;1\;eV^{\alpha} \tag{8}$$

Under the conditions stated above, it can be proved that $E_r^{1+\alpha}\,I_0(\alpha)\;1\;eV^{-\alpha}$ is nearly independent of α, or: $E_r^{1+\alpha}\,I_0(\alpha) = E_r\,I_0\;1\;eV^{\alpha}$, leading to

$$E_r^{-\alpha} = \frac{I_0(\alpha)}{I_0}\;1\;eV^{\alpha} \tag{9}$$

This means that $\Phi_e(E_r)\,E_r$ from Eq. (7) can be calculated as

$$\Phi_e(E_r)\,E_r = \frac{\Phi_e\,I_0(\alpha)}{I_0} \tag{10}$$

or

$$\Phi_e(E_r)\,E_r = \frac{R_e}{I_0} \tag{11}$$

where R_e – epicadmium reaction rate per nucleus:

$$R_e = \frac{A_{sp,e}\, M}{\Theta\, N_A\, \gamma\, \epsilon_p} \quad \text{(in the case of Ge(Li) gamma-counting)} \qquad (12)$$

with

$$A_{sp} = \frac{A_p}{w\, SDC}$$

where A_p – measured average activity of the full-energy peak $= N_p/t_m$; N_p: net number of counts under photopeak collected during measuring time t_m;

w – weight of irradiated element;

S – $1 - e^{-\lambda t_{irr}}$; λ = decay constant, t_{irr} = irradiation time;

D – $e^{-\lambda t_d}$; t_d = decay time;

$$C = \frac{1 - e^{-\lambda t_m}}{\lambda t_m}$$

M – atomic weight;

Θ – isotopic abundance;

N_A – Avogadro's number;

γ – absolute gamma-emission intensity;

ϵ_p – full-energy peak detection efficiency.

Thus, in practice, the method used by SCHUMANN and ALBERT essentially consists in plotting, for a series of well-chosen resonance detectors, log R_e/I_0 versus log E_r, resulting in a straight line with slope $-\alpha$. As the authors state, and as it is obvious from the above discussion the accuracy of the method will depend on the choice of suitable resonance detectors, with respect to the dominance of one single resonance peak, superimposed on a negligible 1/v-part (I_0/σ_0 high). In this context, it is questionable whether the use of ^{55}Mn ($I_0/\sigma_0 = 1.05$) and ^{139}La ($I_0/\sigma_0 = 1.24$), as applied in the method (among others: ^{115}In, ^{197}Au, ^{152}Sm, ^{186}W and ^{98}Mo), can be regarded as appropriate. Moreover, for ^{98}Mo, the dominant resonance energy is given as 480 eV,[8] whereas recent literature data mention almost equally strong resonance peaks at 12.1 eV and 467.4 eV, besides others.[9] At first sight, this situation renders the use of ^{98}Mo as a resonance detector practically impossible.

So as to overcome these difficulties, it seems interesting to generalize the SCHUMANN and ALBERT technique, or, more exactly, to eliminate the need for resonance detectors with one single, dominant resonance peak on a negligible 1/v-tail. This can be accomplished by introducing the concept of the effective reson-

Table 2
Nuclear data for the detectors

Detector	Form used	M	Target isotope	Θ, %	σ_0, barn	I_0, barn	$\frac{I_0}{\sigma_0}$
In	0.1% In Al-wire	114.82	^{115}In	95.7	161	2600	16.1
Au	0.5% Au Al-wire	196.9665	^{197}Au	100	98.8	1550	15.7
Sm	0.05% Sm Al-wire	150.4	^{152}Sm	26.6	206	2900	14.1
W	0.5% W Al-wire	183.85	^{186}W	28.6	38	500	13.8
Mo	0.025 mm Mo-foil	95.94	^{98}Mo	24.1	0.13	6.6	53.8
Zr	0.127 mm Zr-foil	91.22	^{96}Zr	2.8	0.020	5.0	250
			^{94}Zr	17.5	0.052	0.300	5.77

ance energy $\overline{E_r}$ as defined by RYVES.[3,4] Eq. (6), written in terms of $\overline{E_r}$, leads to:

$$\Phi_e(\overline{E_r})\,\overline{E_r} = \Phi_e\,\overline{E_r}^{-\alpha}\ 1\ \text{eV}^\alpha \tag{13}$$

$$= \frac{\Phi_e\,I_0(\alpha)}{I_0(\alpha)}\,\overline{E_r}^{-\alpha}\ 1\ \text{eV}^\alpha \tag{14}$$

Substituting Eq. (5) into Eq. (14), and replacing $\Phi_e\,I_0(\alpha)$ by R_e gives finally:

$$\Phi_e(\overline{E_r})\,\overline{E_r} = \frac{R_e}{I_0 - 0.426\,\sigma_0 + \dfrac{(\overline{E_r})^\alpha\,0.426\,\sigma_0}{(2\alpha+1)\,E_{Cd}^\alpha}} \tag{15}$$

[See note under Eq. (5).]

used for α-determination

Main resonance energy, E_r, eV	Effective resonance energy, $\overline{E_r}^4$, eV	Isotope formed	Half-life	Gamma-energy, $E\gamma$, keV	γ, %
1.457	1.51	^{116m}In	54.2 m	416.9	30.0
4.906	5.47	^{198}Au	2.697 d	411.8	95.53
8.047	8.33	^{153}Sm	46.5 h	103.2	28.2
18.84	19.5	^{187}W	23.9 h	479.5 685.7	26.6 33.0
12.1 467.4	211	^{99}Mo	66.02 h	181.1 739.5	6.0 12.6
302	340	^{97}Zr	16.9 h	743.4	94.2
2263 5800	4520	^{95}Zr	63.98 d	724.2 756.7	44.33 54.52

Eq. (15) replaces Eq. (11) for the calculation of $\Phi_e(\overline{E_r})\,\overline{E_r}$. Since the expression for computing $\Phi_e(\overline{E_r})\,\overline{E_r}$ from the experimentally measured epithermal reaction rate [right-hand side of Eq. (15)] is now itself a function of α, an iterative procedure should be applied. As a first approximation, log $\Phi_e(\overline{E_r})\,\overline{E_r}$-values for the detectors under consideration, as calculated from Eq. (15) for $\alpha = 0$, are plotted versus log $\overline{E_r}$. The slope of the straight line thus obtained $(-\alpha_1)$ yields α_1, which can then be substituted into Eq. (15) for a second approximation, and so on. This procedure results in quickly converging α_n-values. Effective resonance energies can be calculated according to the relation[3,4] (for negligible epithermal self-shielding $G_r = 1$):

$$\ln \bar{E}_r = \frac{\sum_i \frac{\sigma_i \Gamma_{\gamma,i} \ln E_{r,i}}{E_{r,i}}}{\sum_i \frac{\sigma_i \Gamma_{\gamma,i}}{E_{r,i}}} \tag{16}$$

where i – ith resonance;
σ – total neutron capture cross-section at the maximum of the resonance with energy E_r;
Γ_γ – radiative width.

$\bar{E}_r$-values for the isotopes of interest are listed in Table 2.

Modification of the two-detector method of RYVES[3]

The original RYVES method is based on the following equation, written in the WESTCOTT-convention[10] (replacing the symbol β by α):

$$\Omega(1/2) = \frac{\sigma_{0_1}\left[\left(\frac{2g}{2\alpha+1}\right)\left(\frac{E_0}{E_{Cd}}\right)^{\alpha+1/2} - W' + FG_r h(\alpha, G_r) I'/\sigma_0\right]_1}{\sigma_{0_2}\left[\left(\frac{2g}{2\alpha+1}\right)\left(\frac{E_0}{E_{Cd}}\right)^{\alpha+1/2} - W' + FG_r h(\alpha, G_r) I'/\sigma_0\right]_2} \tag{17}$$

where W' – correction factor for non $-1/v$ below E_{Cd};
G_r – epithermal self-shielding factor;
F – Cd-filter epithermal neutron transmission factor;
$h(\alpha, G_r) = \left(\frac{E_0}{\bar{E}_r}\right)^\alpha$
I' – reduced resonance integral (integrated from E_{Cd});
g – WESTCOTT factor[10];
$\Omega(1/2)$ – ratio of the epicadmium reaction rate per nucleus [Eq. (12)] of two detectors 1 and 2.

$$\Omega(1/2) = \frac{\dfrac{A_{sp,e(1)}\,M_1}{\Theta_1\,N_A\,\gamma_1\,\epsilon_{p1}}}{\dfrac{A_{sp,e(2)}\,M_2}{\Theta_2\,N_A\,\gamma_2\,\epsilon_{p2}}} \tag{18}$$

The original method makes use of ^{56}Mn and ^{198}Au as detectors, and consists in plotting graphically the right-hand side of Eq. (17) as a function of α, and reading off α from $\Omega\,(^{56}Mn/^{198}Au)$ found from the cadmium-covered activities of Mn and Au-foils of standard thickness, for which all the above mentioned correction factors are tabulated.

Obviously, the method can be applied using other detector couples with appropriate and known characteristics and nuclear parameters. However, since only two detectors are used, the accuracy of the method will be greatly dependent on the accuracy of the absolute nuclear data to be used in Eq. (17). This is especially true for σ_0 and γ-values, since it has been demonstrated that in most cases these data show a large scattering in the literature.[6,7] Although in principle the same remark holds for the multiresonance – detector method [Eq. (15)], it may be expected that in this technique the use of a large number of detectors will reduce or eventually compensate the effect of systematic errors in the individual absolute nuclear data.

In order to eliminate the use of absolute nuclear data, the method of RYVES can be modified by the introduction of Cd-ratio measurements. Eq. (18) can be transformed into:

$$\Omega(1/2) = \frac{\dfrac{A_{sp,e(1)}\,M_1}{\Theta_1\,N_A\,\gamma_1\,\epsilon_{p1}\,\sigma_{0,1}\,\Phi_s}}{\dfrac{A_{sp,e(2)}\,M_2}{\Theta_2\,N_A\,\gamma_2\,\epsilon_{p2}\,\sigma_{0,2}\,\Phi_s}} \cdot \frac{\sigma_{0,1}}{\sigma_{0,2}} \tag{19}$$

with Φ_s – subcadmium neutron flux $= v_0 n_s$ (as it appears in the HØGDAHL convention[5]);

n_s – subcadmium neutron density;

$$= \int_0^{E_{Cd}} n(v)\,dv$$

Since

$$\frac{\Theta N_A \gamma \epsilon_p \sigma_0 \Phi_s}{M} = A_{sp} - A_{sp,e} \tag{20}$$

where A_{sp} and $A_{sp,e}$ denote the specific activities obtained after a bare and Cd-covered irradiation, respectively, and from the definition of the cadmium ratio:

$$R_{Cd} = \frac{A_{sp}}{A_{sp,e}} \tag{21}$$

Eq. (19) can be reduced to:

$$\Omega(1/2) = \frac{(R_{Cd} - 1)_2}{(R_{Cd} - 1)_1} \cdot \frac{\sigma_{0,1}}{\sigma_{0,2}} \tag{22}$$

When combining Eq. (22) with Eq. (17), one obtains, after introducing $I'/\sigma_0 = I_0/\sigma_0 - 0.426$, and omitting small correction factors [e.g. assuming a 1/v-dependence up to 1–2 eV ($g = 1$, $W' = 0$), using sufficiently diluted detectors ($G_r = 1$) and neglecting epithermal neutron shielding by the Cd-filter ($F = 1$)]:

$$\frac{(R_{Cd} - 1)_2}{(R_{Cd} - 1)_1} = \frac{\left[\left(\frac{I_0}{\sigma_0}\right)_1 - 0.426\right] (E_{r,1})^{-\alpha} + \frac{0.426}{(2\alpha + 1) E_{Cd}^{\alpha}}}{\left[\left(\frac{I_0}{\sigma_0}\right)_2 - 0.426\right] (E_{r,2})^{-\alpha} + \frac{0.426}{(2\alpha + 1) E_{Cd}^{\alpha}}} \tag{23}$$

[See note under Eq. (5).]

From this equation, α can be solved after the measurement of the Cd-ratio for two suitable detectors, without the use of absolute nuclear data or without knowledge of the detection efficiency curve of the Ge(Li) detector.

It should be mentioned that Eq. (23) can also be easily derived from the well-known relation:

$$\frac{(R_{Cd} - 1)_2}{(R_{Cd} - 1)_1} = \frac{\frac{\Phi_s}{\Phi_e} \Big/ \left(\frac{I_0(\alpha)}{\sigma_0}\right)_2}{\frac{\Phi_s}{\Phi_e} \Big/ \left(\frac{I_0(\alpha)}{\sigma_0}\right)_1} \tag{24}$$

After replacing $I_0(\alpha)$ by expression (5), Eq. (23) is obtained.

Experimental

The above described generalized and modified methods have been applied to the α-determination in channel 15 of the Thetis reactor (Gent). This irradiation position is situated in the graphite reflector at a relatively large distance from the reactor core.[13] For the generalized multi resonance – detector method, use was made of ^{115}In, ^{197}Au, ^{152}Sm, ^{186}W, ^{98}Mo, ^{96}Zr and ^{94}Zr. For the modified RYVES method, the ^{94}Zr/^{197}Au detector couple was applied. Although in this case principally also the combinations ^{96}Zr/^{197}Au and ^{94}Zr/^{96}Zr could be used, since Au–Al wire was coirradiated with Zr-foil, these results were not taken into consideration because of the much larger error propagation factors, originating from the Cd-ratio very near to one of ^{96}Zr and from the smaller spread on the respective $\overline{E}_r$-values.

The relevant nuclear data for the isotopes under consideration are summarized in Table 2. For comparison, the main resonance energy, which in the case of ^{98}Mo and ^{94}Zr deviates considerably from the effective resonance energy, has been included.

For the calculations in the multi resonance – detector method, a correction of 9% was made for the Cd neutron transmission factor in the case of ^{186}W, since there is an overlap of a ^{113}Cd resonance at 18.5 eV with the ^{186}W resonance at 18.8 eV.[11]

All detector materials were irradiated in sufficiently diluted form or as thin foils (Mo, Zr) to avoid neutron self-shielding effects. Countings were performed at 15 cm distance on a 70 cm^3 coaxial Ge(Li) detector, coupled to a 4000-channel analyser. All experiments were carried out in triplicates.

The results of the generalized multi resonance – detector method is represented in Fig. 1, showing the final log $\Phi_e(\overline{E}_r)\,\overline{E}_r$ vs. log $\overline{E}_r$ line, obtained after a three-step iteration. It can be observed that all points fall on a straight line. Least-square regression analysis yields an α-value of 0.084 ± 0.011. For comparison, Fig. 1 demonstrates also the points for ^{98}Mo and ^{94}Zr, obtained when applying the original procedure with the use of the main resonance energies from Table 2 (taking the formerly reported literature data of 480 eV for ^{98}Mo[8] and 2265 eV for ^{94}Zr[12]). These points do not fit all to the straight line.

As a result of the modified RYVES method, using Cd-ratio measurements for ^{94}Zr and ^{197}Au, an α-value of 0.084 ± 0.006 is obtained, which is in excellent correspondence with the one obtained from the generalized multi resonance – detector method.

It is interesting to note that the generalized multi resonance – detector method, as proposed in this work, gives sufficiently accurate result when using only the three detectors ^{197}Au, ^{96}Zr and ^{94}Zr, which a suitable spread on their effective resonance energies, namely 5.47 eV, 340 eV and 4520 eV, respectively. From the points in Fig. 1 for these isotopes, an α-value of 0.093 ± 0.018 is obtained,

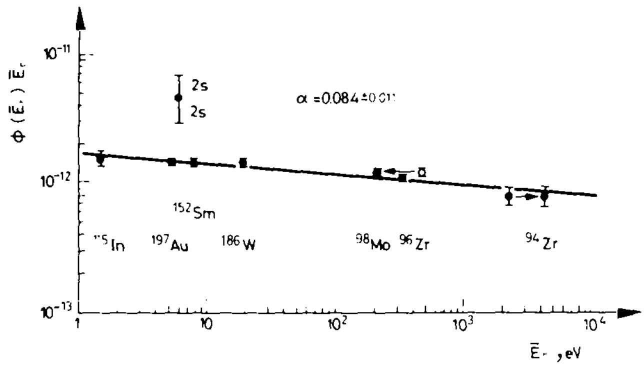

Fig. 1. Determination of α in CH. 15 by the multi resonance detector method

whereas an independent additional experiment yielded 0.084 ± 0.010. These results are in good agreement with the result obtained from all the detector points (0.084 ± 0.011). This enables α to be determined from the generalized multi resonance – detector method by a simple coirradiation, under Cd-cover, of Zr-foil and of sufficiently diluted Au–Al wire.

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