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Data Section

THE EFFECTIVE RESONANCE ENERGY $\bar{E}_r$ AS A PARAMETER FOR THE CORRECTION OF RESONANCE INTEGRALS IN $1/E^{1+\alpha}$ EPITHERMAL NEUTRON SPECTRA; TABULATION OF $\bar{E}_r$-VALUES FOR 96 ISOTOPES

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The present paper shows that the effective resonance energy $\bar{E}_r$, introduced by RYVES, is a useful parameter for the correction of resonance integrals in non-ideal epithermal neutron spectra, which can be approximated by a $1/E^{1+\alpha}$-flux distribution. The definition, the characteristics and the calculation of $\bar{E}_r$ are discussed thoroughly. Tabulations are included, giving $\bar{E}_r$-values for 96 isotopes.

Introduction

In "ideal" reactors and isotopic neutron sources the epithermal neutron flux is accepted to be inversely proportional to the neutron energy.[1] However, in actual irradiation sites, epithermal neutron spectra, deviating from this ideal 1/E-shape, can be expected.[2–8] It was shown that these deviating spectra approximately follow a $1/E^{1+\alpha}$-distribution ($\alpha \gtrless 0$).[4,6]

In many activation analytical techniques, epithermal neutrons give rise to an important fraction or even the full produced activity. Where this is the case, and when in addition absolute or single comparator techniques, using resonance integrals, are applied, the effect of the non-ideality of the epithermal spectrum should not be underestimated.

In the following it is demonstrated how the deviation from the 1/E-law influences the epithermal – more explicitely the epicadmium – reaction rate.

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Ideal 1/E-epithermal neutron spectrum	Deviating $1/E^{1+\alpha}$-epithermal neutron spectrum

$$R_e = \int_{E_{Cd}}^{\infty} \Phi_e(E)\,\sigma(E)\,dE \qquad (1)$$

since: $\Phi_e(E) = \frac{\Phi_e(E_a) \cdot E_a}{E}$	since: $\Phi_e(E) = \frac{\Phi_e(E_a) \cdot E_a^{1+\alpha}}{E^{1+\alpha}}$

where E_a is an arbitrary energy, for simplicity chosen as 1 eV

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

and, per definition, $\Phi_e = \Phi_e(1\text{ eV}) \cdot 1\text{ eV}$

$\Phi_e(E) = \Phi_e \cdot \frac{1}{E}$	$\Phi_e(E) = \Phi_e \cdot \frac{1\text{ eV}^{\alpha}}{E^{1+\alpha}}$
thus: $R_e = \Phi_e \int_{E_{Cd}}^{\infty} \frac{\sigma(E)dE}{E} \qquad (2)$ $= \Phi_e I_0$	thus: $R_e = \Phi_e \int_{E_{Cd}}^{\infty} \frac{\sigma(E)\ 1\text{ eV}^{\alpha}}{E^{1+\alpha}}\,dE \qquad (2')$ $= \Phi_e I_0(\alpha)$
with: $I_0 = \int_{E_{Cd}}^{\infty} \frac{\sigma(E)dE}{E} \qquad (3)$	with: $I_0(\alpha) = \int_{E_{Cd}}^{\infty} \frac{\sigma(E)\ 1\text{ eV}^{\alpha}}{E^{1+\alpha}}\,dE \qquad (3')$

where R_e – epicadmium reaction rate per nucleus;

$\Phi_e(E)$ – epithermal flux per unit of neutron energy interval;

$\sigma(E)$ – (n, γ)-cross-section;

E_{Cd} – effective Cd cut-off energy
= 0.55 eV for a neutron absorber with $\sigma(v) \sim 1/v$ up to 1–2 eV, irradiated in an isotropic neutron flux as a small sample in a cylindrical Cd-box with 1 mm wall thickness (height/dia. = 2);[9]

$I_0, I_0(\alpha)$ – infinite dilution resonance integral for a 1/E and $1/E^{1+\alpha}$ spectrum, respectively;

Φ_e – energy-independent proportionality constant
= integrated epithermal flux per unit of ln(E)-interval (for a 1/E-spectrum) or per unit of $E^{-\alpha}/-\alpha$-interval (for a $1/E^{1+\alpha}$-spectrum).

It is important to notice that the activity produced by non-ideally distributed epithermal neutrons corresponds to Eq. (2') instead of Eq. (2). By consequence, literature data for I_0, which are valid only for an ideal spectrum [Eq. (3)], are not valid in deviating $1/E^{1+\alpha}$-spectra. Instead, $I_0(\alpha)$-values [Eq. (3')] must be used, which fortunately can be calculated from literature I_0-values. For this conversion, the effective resonance energy $\overline{E_r}$ must be used.

The concept of the effective resonance energy has been introduced by RYVES et al.[6,10] Since in these papers the discussion on this subject was rather concise, the present work includes a more extensive description on the definition and characteristics of $\overline{E_r}$.

Definition of $\overline{E}_r$; conversion of I_0 to $I_0(\alpha)$

The effective resonance energy is defined as:

$$(\overline{E_r})^{-\alpha} = \frac{I'_0(\alpha)}{I'_0}\,1\ \text{eV}^{-\alpha} \tag{4}$$

In Eq. (4), I'_0 and $I'_0(\alpha)$ are the reduced resonance integrals (1/v-tail subtracted) for a 1/E and a $1/E^{1+\alpha}$-spectrum respectively.

$$I'_0 = \int_{E_{Cd}}^{\infty}\left[\sigma(E) - \frac{\sigma_0 v_0}{v}\right]\frac{dE}{E} \tag{5}$$

$$= I_0 - \int_{E_{Cd}}^{\infty}\frac{\sigma_0 v_0}{v}\,\frac{dE}{E}$$

$$= I_0 - 0.426\ \sigma_0 \tag{6}$$

and

$$I'_0(\alpha) = \int_{E_{Cd}}^{\infty}\left[\sigma(E) - \frac{\sigma_0 v_0}{v}\right]1\ \text{eV}^{-\alpha}\frac{dE}{E^{1+\alpha}} \tag{5'}$$

$$= I_0(\alpha) - \int_{E_{Cd}}^{\infty}\frac{\sigma_0 v_0}{v}\,1\ \text{eV}^{-\alpha}\frac{dE}{E^{1+\alpha}}$$

$$= I_0(\alpha) - \sigma_0\,\frac{1\ \text{eV}^{-\alpha}\sqrt{E_0}}{(\alpha + 1/2)\,E_{Cd}^{\alpha+1/2}}$$

$$= I_0(\alpha) - \sigma_0\,\frac{0.426\ 1\ \text{eV}^{\alpha}}{(2\alpha + 1)E_{Cd}^{\alpha}} \tag{6'}$$

where v – neutron velocity;
v_0 – 2200 $m \cdot s^{-1}$;
$E_0 = 1/2\ m\ v_0^2 = 0.025$ eV;
$\sigma_0 = \sigma(E_0)$

It should be remarked that the definition of $\bar{E}_r$, expressed by Eq. (4), differs slightly from the definition given by RYVES. Indeed, in the present work, resonances below E_{Cd} are not taken into account when calculating I_0' and $I_0'(\alpha)$, whereas in the original papers,[6,10] the resonances between μkT and E_{Cd} are included as well (where $\mu = 5$, k = Boltzmann's constant, T = the temperature corresponding to the maximum of the Maxwellian neutron flux distribution; for T = 293.15 K, $\mu kT = 0.125$ eV). However both definitions are meaningful and for most isotopes are even equivalent since only a few isotopes show reasonance peaks in the interval under consideration.

When introducing Eq. (6) and Eq. (6') into Eq. (4), one obtains after rearrangement:

$$I_0(\alpha) = \left(\frac{I_0 - 0.426\ \sigma_0}{(\bar{E}_r)^\alpha} + \frac{0.426\ \sigma_0}{(2\alpha + 1)\, E_{Cd}^\alpha} \right) \tag{7}$$

This equation makes it possible to convert I_0 to $I_0(\alpha)$, if $\bar{E}_r$ and α are known.

Eqs (6), (6'), and (7) are only valid for $E_{Cd} = 0.55$ eV, since $0.426 = 2 \cdot (E_0/E_{Cd})^{1/2}$ when $E_0 = 0.025$ eV and $E_{Cd} = 0.55$ eV.

The influence of α on the resonance integrals is demonstrated (Table 1) listing $I_0(\alpha)$ for $\alpha = 0.1$, a value which may occur at relatively large distances from the core in a small, graphite moderated reactor (6). Comparing $I_0(\alpha = 0.1)$ to $I_0(\alpha = 0)$, differences are occurring as a factor 1.18 for ^{197}Au, 1.79 for ^{96}Zr and 2.16 for ^{94}Zr.

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 $\bar{E}_r$ eV	σ_0 barn	1/E-spectrum I_0 ($\alpha = 0$), barn	$1/E^{1+\alpha}$-spectrum I_0 ($\alpha = 0.1$); Eq. (7), 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

Experimental determination of α

Different procedures for the determination of α, as well as some experimental results were discussed elsewhere.[2-8] However, so as to give a rough idea about the extent of the non-ideality of the epithermal spectrum in a real irradiation site, it can be mentioned that in the small, graphite-moderated Thetis reactor α-values were found ranging from –0.028 in the core, up to 0.11 when moving away from the core in the graphite.[11]

Calculation of $\overline{E}_r$

If in the definition of $\overline{E}_r$ [Eq. (4)], I_0' and $I_0'(\alpha)$ are replaced by the summation of the integrated Breit–Wigner expressions for all resonance s above E_{Cd}, one obtains:

$$(\overline{E}_r)^{-\alpha} = \frac{\sum\limits_i \frac{\pi}{2} \frac{\sigma_i \Gamma_{\gamma,i}}{E_{r,i}} E_{r,i}^{-\alpha}}{\sum\limits_i \frac{\pi}{2} \frac{\sigma_i \Gamma_{\gamma,i}}{E_{r,i}}} \tag{8}$$

where i – refers to the i-th resonance;
σ – total neutron capture cross-section at the maximum of the resonance at energy E_r;
Γ_γ – radiation width.

For these integrations of the Breit-Wigner equations, it was assumed that the resonance peaks are narrow, i. e. $\Gamma_i \ll E_{r,i}$ (Γ_i = total width).[16] In this way, E can be taken constant and equal to $E_{r,i}$ for each resonance peak. The errors, introduced by this approximation, amount to a few percent in both nominator and denominator of Eq. (8), so that partial cancelling can be expected. In addition, in the conversion of I_0 to $I_0(\alpha)$ [Eq. (7)], the error on $\overline{E}_r$ is considerably reduced, as will be demonstrated further on. It is assumed in Eq. (8) that resonance self-shielding is negligible, which certainly is the case when using sufficiently diluted or thin detectors.

From Eq. (8) it follows that the effective resonance energy $\overline{E}_r$ is a function of α, and thus in principle, is no real nuclear constant for a given isotope.

When expanding the $(E_r)^{-\alpha}$ factors in series, it is possible however, to omit the higher order terms for sufficiently low $E_{r,i}$, E_r and/or α-values. Thus:

$$(E_r)^{-\alpha} = e^{-\alpha \ln E_r} = 1 - \alpha \ln\left(E_r + \frac{\alpha^2}{2!}(\ln E_r)^2 - \ldots\right) \tag{9}$$

Introducing expression 9 for $\bar{E}_r$ and $E_{r,i}$ into Eq. (8) gives, after some rearrangement:

$$\ln \bar{E}_r = \frac{\sum\limits_i \dfrac{\sigma_i \Gamma_{\gamma,i} \ln E_{r,i}}{E_{r,i}}}{\sum\limits_i \dfrac{\sigma_i \Gamma_{\gamma,i}}{E_{r,i}}} \tag{10}$$

Eq. (10) yields $\bar{E}_r$ independent of α, but due to the introduced approximations, it is, strictly speaking, only valid for low $\bar{E}_r$ and/or α-values. However, since the expansion into series is performed on both the left and right-hand terms of Eq. (8), it seems likely that the above mentioned approximations will not lead to serious differences in the $\bar{E}_r$ values calculated from Eq. (8) (for a given α) or from Eq. (10), even for high $\bar{E}_r$ and α-values. This can be checked numerically with some practical examples. Table 2 summarizes the results for ^{197}Au, ^{96}Zr and ^{94}Zr, comparing the approximate $\bar{E}_r$-values [Eq. (10] with the exact $\bar{E}_r$-values [Eq. (8)] for α factors of 0.01 and 0.1.

Table 2
Comparison of approximate and exact $\bar{E}_r$-values

Target isotope	$\bar{E}_r$[Eq.(8)] for $\alpha = 0.01$, eV	$\bar{E}_r$[Eq.(8)] for $\alpha = 0.1$, eV	Approximate $\bar{E}_r$ [Eq.(10)], eV
^{197}Au	5.47	5.40	5.47
^{96}Zr	339	335	340
^{94}Zr	4513	4451	4520

From Table 2 it appears that, even with such a high resonance energy as for ^{94}Zr and with such a high α-factor as 0.1, the difference between the approximate and exact $\bar{E}_r$-value is not more than 1.6%.

This means that, with a good approximation, in all cases $\bar{E}_r$-values can be calculated from the resonance parameters according to Eq. (10). Thus they can be tabulated as nuclear constants, nearly independent of the epithermal neutron flux distribution, and valid when resonance self-shielding is negligible.

Physical meaning of the effective resonance energy

RYVES concluded before: "The significance of this energy $\overline{E_r}$ is, that it represents a single effective average energy, which replaces all the resonance energies $E_{r,i}$."[6]

Indeed Eq. (10) shows that ln $\overline{E_r}$ is the weighted mean of the separate ln $E_{r,i}$ values, with the resonance peak areas, $\pi/2\ \sigma_i\ \Gamma_{\gamma,i}$ ($\pi/2$ cancels in nominator and denominator), and the inverse of the resonance energies, $1/E_{r,i}$, as weighing factors.

Tabulation of $\overline{E_r}$

For the calculation of $\overline{E_r}$ [Eq. (10)] the resonance parameter data can be taken from BNL-325[12,13] or from the NEA Data Bank.

For each isotope all resonance peaks were taken into account for which the necessary data could be found. In case that, for a given isotope, $\Gamma_{\gamma,i}$-values are not available for the smaller resonance peaks, the $\Gamma_{\gamma,i}$ for the main resonance can be adopted. Indeed, the radiation widths in a single nucleus are known to be essentially constant from level to level.[14] Lacking σ_i-values can be calculated from additional resonance parameters, according to Eq. (4), page xii of BNL-325.[12]

Table 3 summarizes calculated $\overline{E_r}$-values for 96 isotopes. It should be noted that no isotopes having resonance energies lower than 1 eV were included in this work. This limitation originates from the given definition of $\overline{E_r}$ [Eqs (4), (5) and (5')] using epicadmium resonance integrals and from the definition of the effective Cd cut-off energy (= 0.55 eV only valid for 1/v detectors up to 1–2 eV).

Mostly negligible errors may have been introduced due to the incomplete knowledge of resonance parameters (e.g. s- or p-wave resonances, unresolved resonances, etc.). However, it should be noted that in the calculation of $I_0(\alpha)$ according to Eq. (7), a considerable error reduction factor appears concerning the error on $\overline{E_r}$; when e.g. for ^{96}Zr a $\overline{E_r}$-value of 300 eV instead of the correct 340 eV (i.e. 12% error) would be used, this would produce an error on the I_0 ($\alpha = 0.1$) value of only 1.3% [$I_0(\alpha = 0.1) = 2.83$ instead of 2.79].

Table 3
Effective resonance energy

Target isotope	$\overline{E_r}$, eV	Target isotope	$\overline{E_r}$, eV	Target Isotope	$\overline{E_r}$, eV
^{23}Na	3 130	^{89}Y	3 440	^{140}Ce	11 400
^{26}Mg	220 000	^{94}Zr	4 520	^{141}Pr	251
^{27}Al	8 240	^{96}Zr	340	^{146}Nd	728
^{37}Cl	30 600	^{93}Nb	184	^{148}Nd	202
^{41}K	9 040	^{98}Mo	211	^{150}Nd	153
^{45}Sc	2 120	^{100}Mo	513	^{152}Sm	8.33
^{51}V	5 960	^{102}Ru	717	^{154}Sm	136
^{50}Cr	5 950	^{104}Ru	347	^{153}Eu	3.29
^{55}Mn	412	^{103}Rh	1.43	^{158}Gd	45.4
^{58}Fe	325	^{108}Pd	37.9	^{160}Gd	436
^{59}Co	133	^{107}Ag	31.4	^{159}Tb	11.4
^{64}Ni	14 300	^{109}Ag	14.1	^{164}Dy	145
^{63}Cu	742	^{110}Cd	114	^{165}Ho	9.73
^{65}Cu	452	^{114}Cd	178	^{169}Tm	4.77
^{64}Zn	428	^{116}Cd	576	^{174}Yb	390
^{68}Zn	516	^{113}In	9.81	^{176}Yb	389
^{69}Ga	192	^{115}In	1.51	^{175}Lu	8.47
^{71}Ga	152	^{112}Sn	111	^{176}Hf	12.3
^{74}Ge	2 940	^{116}Sn	136	^{181}Ta	7.61
^{76}Ge	582	^{122}Sn	289	^{186}W	19.5
^{75}As	102	^{124}Sn	64.6	^{185}Re	2.54
^{74}Se	29.5	^{121}Sb	11.2	^{187}Re	12.0
^{78}Se	419	^{123}Sb	24.9	^{193}Ir	1.46
^{80}Se	3 550	^{122}Te	83.2	^{194}Pt	418
^{82}Se	8 540	^{126}Te	207	^{196}Pt	565
^{79}Br	51.4	^{128}Te	508	^{198}Pt	95.8
^{81}Br	114	^{130}Te	3 230	^{197}Au	5.47
^{85}Rb	694	^{127}I	43.0	^{196}Hg	93.5
^{87}Rb	376	^{133}Cs	8.68	^{198}Hg	41.5
^{84}Sr	464	^{134}Ba	102	^{202}Hg	1 960
^{86}Sr	672	^{138}Ba	20 400	^{232}Th	53.5
^{88}Sr	15 500	^{139}La	76.8	^{238}U	15.8

Conclusion

The present work shows that for several (n, γ) activation techniques, in which epithermal activation is concerned, the effective resonance energy should be considered as an indispensable parameter when a good accuracy is required.

Several of the listed $\overline{E}_r$-values were already applied in the single-comparator method based on k_0-factors.[15]

In the future these tabulations will be completed and the present values might be the subject of further refinement. In cases where the necessary parameters for the calculation of $\overline{E}_r$ [Eq. (10)] are not available in literature, the experimental determination of $\overline{E}_r$ is considered.

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