

THE EFFECTIVE RESONANCE ENERGY AS A PARAMETER IN
(n, γ) ACTIVATION ANALYSIS WITH REACTOR NEUTRONS

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(Received December 12, 1986)

In absolute or comparator standardization methods of (n, γ) activation analysis with reactor neutrons, the epithermal neutron flux nonideality, if neglected, may lead to significant errors on the analytical result. The concept of the effective resonance energy (\bar{E}_r), introduced to correct for this effect, is reviewed.

Introduction

In thermal reactor theory it is usually accepted that the epithermal neutron flux per unit energy interval is inversely proportional to the neutron energy $1/E$:

$$\phi'_e(E) = \frac{\phi_e}{E} \quad (1)$$

with ϕ_e = the conventional "epithermal flux". Eq. (1) is derived from a simplified model of the moderation process, where the following assumptions are introduced:

- moderation takes place in a homogeneous, infinite medium;
- the sources of fission neutrons (i.e. fuel) are homogeneously distributed throughout the moderator;
- moderator atoms are free and at rest before being struck by neutrons;
- absorption (resonance or $1/v$ absorption), inelastic and anisotropic elastic scattering during moderation do not occur;
- elastic scattering is energy independent.

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The resonance integral (I_0), an essential nuclear parameter when applying absolute or comparator (e.g. k_0 - /2/, /3/) standardization methods in (n, γ) activation analysis with reactor neutrons (NAA), is defined, measured, tabulated in literature and should be used assuming such "ideal" $(1/E)$ epithermal flux distribution:

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

where E_{Cd} (=0.55 eV) is the effective Cd cut-off energy and $\sigma(E)$ = the (n, γ) cross-section.

However, the (over)simplifications introduced to describe the moderation process are valid to a different extent from one reactor to another or even within the same reactor /4/. This means that the actual epithermal flux distribution deviates from the ideal $1/E$ -law and is found to be better approximated by a semiempirical form /5/-/9/:

$$\phi'_e(E) = \frac{\phi_e}{E^{1+\alpha}} E_a^\alpha \quad (3)$$

with E_a = an arbitrary energy (usually = 1eV) and α = an experimentally determinable /10/, /11/ characteristic of the reactor site (to a first approximation energy independent).

Consequently the resonance integral should be modified to

$$I_0(\alpha) = \int_{E_{Cd}}^{\infty} \sigma(E) \frac{E_a^\alpha}{E^{1+\alpha}} dE \quad (4)$$

So as to enable the use of the compiled and tabulated I_0 -values in real $(1/E^{1+\alpha})$ circumstances, i.e. to enable the $I_0 \rightarrow I_0(\alpha)$ conversion, the concept of the effective resonance energy (\bar{E}_r) was introduced /6/, /7/, /12/, /13/. \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" /14/ and is defined as an α -dependent parameter :

$$\bar{E}_r(\alpha) \approx E_a \left[\frac{I'_0(\alpha)}{I'_0} \right]^{(-1/\alpha)} \quad (5)$$

with $I'_0(\alpha)$ and I'_0 =reduced resonance integrals (1/v-part subtracted). For $E_a=1\text{eV}$, Eq.(5) gives $\bar{E}_r(\alpha)$ in eV.

The $I_0 \rightarrow I_0(\alpha)$ conversion is: ($\bar{E}_r(\alpha) \rightarrow \bar{E}_r$ transition : see below)

$$I_0(\alpha) = \left[\frac{I_0 - 0.429 \sigma_0}{\bar{E}_r^\alpha} + \frac{0.429 \sigma_0}{(2\alpha+1)E_{Cd}^\alpha} \right] E_a^\alpha \quad (6)$$

where $\sigma_0 = 2200 \text{ m.s}^{-1}$ (n, γ) cross-section. With \bar{E}_r and E_{Cd} in eV, the term $E_a^\alpha = (1\text{eV})^\alpha \approx 1$ can be omitted.

In terms of the closely related and practical Q_0 -factors ($Q_0 = I_0 / \sigma_0$), the above becomes:

$$Q_0(\alpha) = \frac{I_0(\alpha)}{\sigma_0} = \frac{Q_0 - 0.429}{\bar{E}_r^\alpha} + C_\alpha \quad (7)$$

$$\text{with } C_\alpha = \frac{0.429}{(2\alpha+1) 0.55^\alpha} \quad (8)$$

Impact of the epithermal flux nonideality on the analytical result

When applying absolute or comparator type NAA, the concentration of an element in the sample is proportional to a term $f/2$

$$\rho \sim \frac{f + Q_0^*(\alpha)}{f + Q_0(\alpha)} \quad (9)$$

where the asterisk refers to the coirradiated flux monitor and $f = \phi_s / \phi_e$ is the thermal to epithermal flux ratio. It is clear that an error will be made when neglecting the epithermal flux nonideality (by assuming $\alpha=0$), i.e. when taking Q_0 and Q_0^* in place of $Q_0(\alpha)$ and $Q_0^*(\alpha)$. Denoting by ρ' the calculated concentration with α neglected, the relative percentual error is given by

$$s_\rho = \left| \frac{\rho - \rho'}{\rho} \right| \cdot 100 = \left| 1 - \frac{f + Q_0^*}{f + Q_0^*(\alpha)} \cdot \frac{f + Q_0(\alpha)}{f + Q_0} \right| \cdot 100 \quad (10)$$

Having in mind Eq.(7), one can see that the magnitude of the error depends on:

- the isotope, characterized by Q_0 and \bar{E}_r values;
- the irradiation conditions, i.e. f and α ;
- the comparator used (Q_0^x and \bar{E}_r^x);
- the type of analysis, i.e. whether the sample is irradiated in the whole reactor flux (reactor neutron activation analysis), or with the thermal neutrons screened out, usually by means of Cd-filter (epicadmium neutron activation analysis - ENAA), the latter equivalent to $f = \phi_s / \phi_e = 0$.

To obtain a complete picture of the magnitude and variation of the error, Eq. (10) was applied to 127 isotopes of analytical interest /4/. A rather extreme range of f and α values was considered: $f=15$ to 158 , $\alpha = -0.028$ to 0.110 (reactor THETIS, Gent, Belgium). The following could be observed:

- the error increases with the absolute value of α ;
- errors are larger in ENAA (Cd-covered activation) than in RNAA (bare activation), since in ENAA the whole activity is induced by epithermal neutrons, whose nonideal spectrum causes the error, while in RNAA the error is reduced by the thermal activation contribution;
- the error is smaller for the isotopes whose Q_0 and \bar{E}_r values are close to Q_0^x and \bar{E}_r^x of the flux-monitor (e.g. ^{169}Tm : $Q_0 = 16.4$, $\bar{E}_r = 4.80$ eV; ^{197}Au : $Q_0^x = 15.7$, $\bar{E}_r^x = 5.65$ eV);
- depending on the isotope, three groups can be distinguished:
 - a) low Q_0 (e.g. $Q_0 < 1$) isotopes: the error is practically negligible (<1%) in RNAA, while ENAA has no sense (poor epithermal activation) and should not be considered;
 - b) moderate Q_0 (e.g. $1 < Q_0 < 10$): the errors in RNAA reach up to 8%, ENAA is rarely applied - the errors can be as high as 67% (^{208}Pb);
 - c) high Q_0 (e.g. $Q_0 > 10$) isotopes are suitable for both RNAA and ENAA. Errors reach up to 30% in RNAA (mostly 5-15%) and up to ~50% in ENAA (usually >30%).

The above clearly shows that, in order to preserve the accuracy of the analysis, the impact of the epithermal flux non-ideality should be taken into account when calculating the concentration and, if necessary, corrected for.

Propagation of uncertainties in \bar{E}_r -values towards the analytical result

Let us now examine how accurately we need to know an \bar{E}_r value so as to enable a satisfactory α -correction of the analytical result [Eqs.(7) and (9)]. The relevant error propagation function is /4/:

$$Z_{\rho}(\bar{E}_r) = \left| \frac{\partial \rho}{\partial \bar{E}_r} \cdot \frac{\bar{E}_r}{\rho} \right| = \left| \alpha \frac{Q_0(\alpha) - C_{\alpha}}{f + Q_0(\alpha)} \right| \quad (11)$$

and the corresponding error in the analytical result

$$s_{\rho}(\bar{E}_r) = Z_{\rho}(\bar{E}_r) \cdot s_{\bar{E}_r} \quad (12)$$

By setting, for instance, $s_{\rho}(\bar{E}_r) = 1\%$ as a tolerable error in ρ , originating from the uncertainty in \bar{E}_r , we obtain the "acceptable" uncertainty in \bar{E}_r . This calculation was performed for the 127 isotopes mentioned and an extreme range of f and α values. As already pointed out, low Q_0 isotopes do not require an α -correction. For moderate Q_0 's and \bar{E}_r 's known to within a factor of 2 or 3 are sufficiently accurate, while for high Q_0 's ($Q_0 > 10$) \bar{E}_r -uncertainties of $\sim 50\%$ in RNAA and of 10-20% in ENAA are satisfactory.

When making judgements about the required accuracy in \bar{E}_r -values, α -uncertainties should also be kept in mind. Namely, both \bar{E}_r and α are employed to correct for the epithermal non-ideality. It is found that $Z_{\rho}(\alpha)$ factors are, in general, several times higher than $Z_{\rho}(\bar{E}_r)$. Thus, with $s_{\bar{E}_r}$ and s_{α} comparable, s_{ρ} after correction will mainly originate from the uncertainty in α . In such cases, it makes no sense to ask for a more accurate \bar{E}_r if it is surpassed by unavoidable experimental uncertainty in α /11/.

The achievable uncertainties in \bar{E}_r -values (see further) are in most cases satisfactory for NAA needs.

Determination of the effective resonance energies

There are, in principle, two ways to determine an \bar{E}_r - value: calculation from the neutron resonance parameter data and experimental determination.

Calculation

From the definition of effective resonance energy it follows that \bar{E}_r is not a constant, but a function of α . However, $\bar{E}_r(\alpha)$ can be fairly approximated by an α -independent \bar{E}_r -value given as a weighted mean of the logarithms of the resonance energies $E_{r,i}$ /12/:

$$\ln \bar{E}_r = \frac{\sum_i \omega_i \ln E_{r,i}}{\sum_i \omega_i} \quad (13)$$

with the weighing factors

$$\omega_i = \frac{(g \Gamma_\gamma \Gamma_n / \Gamma)_i}{E_{r,i}^2} \quad (14)$$

and Γ_γ =radiative width, Γ_n =neutron width, Γ =total width of the i -th resonance.

In most cases Eq.(13) yields \bar{E}_r -values which do not differ significantly from $\bar{E}_r(\alpha)$ (up to 20% for a few isotopes and high α 's). Nevertheless, a simple compensation formula

$$\bar{E}_r(\alpha) = \bar{E}_r \cdot e^{-p\alpha} \quad (15)$$

with p -values calculated from resonance parameter data /4/, can be used if necessary.

Based on Eq. (13), the first generation of \bar{E}_r -values (for 96 isotopes) was calculated in 1979 /12/. In Table 1 a second generation, including \bar{E}_r 's for 127 isotopes and the corresponding uncertainties, can be found which were calculated from the newest compilations of resonance parameter data /15/, /16/.

Table 1

Calculated \bar{E}_r -values for 127 analytically interesting (n, γ) reactions. Resonance parameter data were taken from Refs (15, 16); *no resonance data available; **no error assignment possible; [†]experimental value (17)

Target isotope	\bar{E}_r, eV	Target isotope	\bar{E}_r, eV	Target isotope	\bar{E}_r, eV	Target isotope	\bar{E}_r, eV
18O	114000 \pm 80000	80Se	2940 \pm 410	121Sb	13.1 \pm 0.5	169Tm	4.80 \pm 0.10
19F	44700 \pm 2200	82Se	8540 \pm **	123Sb	28.2 \pm 1.7	168Yb	0.61 \pm 0.01
23Na	3380 \pm 370	79Br	69.3 \pm 6.2	120Te	*	174Yb	602 \pm 48
26Mg	257000 \pm 33000	81Br	152 \pm 14	122Te	92.3 \pm 3.7	176Yb	412 \pm 21
27Al	11800 \pm 700	85Rb	839 \pm 50	124Te	1210 \pm 100	175Lu	16.1 \pm 0.8
30Si	2280 \pm 10	87Rb	364 \pm 11	126Te	285 \pm 20	174Hf	29.6 \pm 2.1
31P	38500 \pm 6900	84Sr	469 \pm 33	128Te	738 \pm 52	177Hf	2.08 \pm **
36S	*	86Sr	795 \pm 16	130Te	2950 \pm 210	178Hf	8.01 \pm 0.16
37Cl	13700 \pm 1900	89Y	4300 \pm 340	127I	57.6 \pm 2.3	179Hf	16.2 \pm 1.9
40Ar	31000 \pm 5600	94Zr	6260 \pm 250	133Cs	9.27 \pm 1.02	180Hf	115 \pm 7
41K	2960 \pm 210	96Zr	338 \pm 7	130Ba	69.9 \pm 3.5	181Ta	10.4 \pm 0.6
46Ca	*	93Nb	574 \pm 46	132Ba	143 \pm **	182W	9.20 \pm 0.55
48Ca	1330000 \pm **	98Mo	241 \pm 48	134Ba	115 \pm 6	186W	20.5 \pm 0.2
45Sc	5130 \pm 870	100Mo	672 \pm 94	136Ba	545 \pm 38	185Re	3.40 \pm 0.14
50Ti	63200 \pm 2500	96Ru	776 \pm 124 [†]	138Ba	15700 \pm 500	187Re	41.1 \pm 1.6
51V	7230 \pm 290	102Ru	181 \pm 7	139La	76.0 \pm 3.0	189Os	12.3 \pm 0.4
50Cr	7530 \pm 830	104Ru	495 \pm 50	138Ce	*	190Os	114 \pm 2
55Mn	468 \pm 51	103Rh	1.45 \pm 0.01	140Ce	7200 \pm 1300	192Os	89.7 \pm 3.6
58Fe	637 \pm 153	106Pd	282 \pm 6	142Ce	1540 \pm 1850	193Ir	2.21 \pm 0.20
59Co	136 \pm 7	108Pd	39.7 \pm 2.0	141Pr	296 \pm 12	190Pt	27.6 \pm 0.6
64Ni	14200 \pm 1700	110Pd	950 \pm 86	146Nd	874 \pm 52	196Pt	291 \pm 44
63Cu	1040 \pm 50	107Ag	38.5 \pm 1.9	148Nd	236 \pm 14	198Pt	106 \pm 3
65Cu	766 \pm 130	109Ag	6.08 \pm 0.06	150Nd	173 \pm 21	197Au	5.65 \pm 0.40
64Zn	2560 \pm 260	108Cd	243 \pm 24	152Sm	8.53 \pm 0.09	196Hg	93.5 \pm 0.1
68Zn	590 \pm 60	110Cd	125 \pm 16	154Sm	142 \pm 10	198Hg	39.3 \pm 2.8
69Ga	201 \pm 16	114Cd	207 \pm 39	153Eu	5.80 \pm 0.23	202Hg	1960 \pm 160
71Ga	154 \pm 18	116Cd	726 \pm 87	158Gd	48.2 \pm 3.9	204Hg	*
74Ge	3540 \pm 280	113In	6.41 \pm 0.96	160Gd	480 \pm 34	203Tl	276 \pm 28
76Ge	583 \pm 23	115In	1.56 \pm 0.03	159Tb	18.1 \pm 0.9	205Tl	2960 \pm 360
75As	106 \pm 36	112Sn	107 \pm 3	164Dy	224 \pm 11	206Pb	10500 \pm 1200
74Se	29.4 \pm 1.2	116Sn	128 \pm 4	165Ho	12.3 \pm 0.4	208Pb	145000 \pm 4000
76Se	577 \pm 46	122Sn	424 \pm 59	166Er	59.3 \pm 4.2	209Bi	1210 \pm 60
78Se	501 \pm 35	124Sn	74.2 \pm 5.2	170Er	129 \pm 3	232Th	54.4 \pm 0.5
						238U	16.9 \pm 0.2

Since the quantity and quality of the available resonance parameter data continuously improve, the \bar{E}_r -values should be adequately refined and updated.

The specified uncertainties in calculated \bar{E}_r 's (usually 5-20%) are satisfactory for NAA needs, but, unfortunately, do not account for possible incompleteness of the data. Therefore a number of the other criteria were established /4/ to judge the accuracy of a calculated \bar{E}_r .

Experimental determination

For isotopes whose resonance parameter data are incomplete, inaccurate, obsolete or even not known at all, experimental \bar{E}_r -determinations should be considered. Two such methods were developed and applied to a number of isotopes.

The method for simultaneous Q_0 and \bar{E}_r determination /17/ is based on Cd-ratio measurements of the investigated isotope in several reactor channels with a large spread in α -values. The method provides accurate \bar{E}_r and Q_0 values ($s_{\bar{E}_r} \sim 20\%$, $s_{Q_0} \sim 1\%$).

In the " \bar{E}_r -comparator" method /18/, the isotope whose \bar{E}_r -value is to be measured is coirradiated together with a comparator isotope with well known effective resonance energy $\bar{E}_{r,c}$ (e.g. ^{68}Zn : $\bar{E}_{r,c} = 590$ eV). Knowing α in the irradiation site, the unknown \bar{E}_r can be found from $\bar{E}_{r,c}$ and from the Q_0 's and Cd-ratios of the two isotopes. In both methods flux ratios are measured with a co-irradiated monitor (e.g. ^{197}Au).

The method for simultaneous Q_0 and \bar{E}_r determination yields more accurate results, but demands much experimental work and requires 6-7 irradiation channels with a broad range of α -values. The latter requirement, though fulfilled in the reactor THETIS, is not often met within thermal reactors. The \bar{E}_r -comparator method is simpler, but still asks for a channel with a very high α and can be applied only for isotopes whose Q_0 -values are accurately ($s_{Q_0} = 1-2\%$) known in advance.

Conclusion

When studying the impact of the epithermal flux nonideality on the analytical result, the necessity for the correction was demonstrated. The $\phi'_e(E) \sim 1/E^{1+\alpha}$ epithermal spectrum representation, together with the \bar{E}_r -concept, is found satisfactory for this purpose. For most isotopes, sufficiently accurate \bar{E}_r -values, calculated from neutron resonance parameter data or experimentally determined, are available at present.

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The financial support of the NFWO (Belgium) and of the Association for Scientific Activities of Montenegro (Yugoslavia) is highly appreciated.

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