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Defining "k_f-factors" for threshold reactions

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Abstract
The k_0-method (De Corte in The k_0-standardization method: move to the optimization of neutron activation analysis. Habil. Thesis, Ghent University, Belgium, 1987) was developed solely for the use of (n, γ) nuclear reactions in neutron activation analysis. For this, a definition of only the thermal and epi-thermal flux was needed. The fast flux of the fission neutrons was not taken into account although it was considered for primary interferences by De Corte$_0$. The energy distribution of the fission neutrons can be rather well described by a Watt distribution but is reactor dependent. To complicate things, the activation cross-section behaviour is nuclide dependent. In order to incorporate threshold reactions in the k_0-method we propose to use predefined k_f-factors, measuring the fast flux using a Ni-58 monitor, and to introduce an h-factor that accounts for all deviations for a specific reaction and irradiation facility. It is shown, based on data from Verheijke, that there are useful correlations for Ni-58, Ti-47 and Ti-48. Activation cross section functions indicate that there are possible more relations that might allow h-factors to be predicted.

Keywords Neutron activation analysis · k_0-method · Fast neutron spectrum · Threshold reactions · ENDF/B-VIII.0

Introduction

The k_0-method [1] was developed solely for the use of (n, γ) nuclear reactions in neutron activation analysis. For this a definition of only the thermal and epi-thermal flux was needed. The fast flux, originating directly from the fission neutrons, was not considered for (n, γ)-reactions because of the negligible activation probability at high neutron energies. But fast neutrons can cause significant threshold neutron reactions like (n, p), (n, α), (n, n'), (n,2n) and more. Some of these reactions will cause primary first-order interferences [1–3], some will result in analytical useful nuclides [2].

Quantification of interferences and analytical reactions call for average activation cross sections. These were already calculated based on an activation cross section distribution and the neutron energy spectrum U-235 fission neutrons [4–7]. The U-235 neutron spectrum can be rather well described by a Watt or Maxwell distribution.

However, the neutron energy distribution will differ from this, depending on reactor and irradiation position inside the reactor.

Activation by fast neutrons is normally rather small but can in some specific cases be analytically used. More often fast neutrons from a hard spectrum will cause interferences with other analytical nuclides. In most NAA labs correction factors are used to correct for these primary first-order interferences, as was already done in 1962 by Durham [3]. Up to now there is no general solution for the problem that the fast flux distribution has no fixed universal shape. Nevertheless, average cross sections are defined based on a Watt spectrum and given in literature [2–5]. The cross section, weighed over a Watt spectrum, is averaged between zero and infinity. In most papers as well as in the Inter program (ENDF) the denominator in the formula is incorrectly described as being the integral over the neutron flux from the threshold energy to infinity. Uddin [8] unfolded the fast spectrum of the Triga Mark II reactor in Bangladesh and compared with similar result from the Triga reactor in Slovenia.

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Theory

Activation by fast neutrons depends heavily on the cross-section behaviour as a function of neutron energy E and the shape of the neutron flux spectrum. The reaction rate R for reaction a is given by

$$R = \Phi_{f,i}.\overline{\sigma_{f,a,i}} \tag{1}$$

where $\overline{\sigma_{f,a,i}}$ is the threshold neutron capture cross-section $\sigma_a(E)$ averaged over the fast neutron spectrum $\varphi_{f,i}(E)$ in irradiation facility i:

$$\overline{\sigma_{f,a,i}} = \frac{\int_0^\infty \sigma_a(E)\varphi_{f,i}(E)dE}{\int_0^\infty \varphi_{f,i}(E)dE} \tag{2}$$

and

$$\Phi_{f,i} = \int_0^\infty \varphi_{f,i}(E)dE \tag{3}$$

The fast neutron energy spectrum depends on the configuration of the reactor, fuel composition and configuration around the irradiation channel. To standardise this, typically the U-235 fission flux distribution, described by a Watt distribution, is often used to calculate average cross-sections. There are different functions and integration limits in use for the calculations [Erdtmann, ENDF (Inter), empirical relations for U-235 and Pu-239].

The Watt distribution is given by

$$\varphi_f(E) = Ce^{-E/a}\sinh\left(\sqrt{bE}\right) \tag{4}$$

where a, b and C are fission-reaction dependent parameters. The Maxwell distribution is given by

$$\varphi_f(E) = Ce^{-E/a}\sqrt{E} \tag{5}$$

where a is a fission-reaction dependent parameter. For both distributions, the energy of the incoming neutrons (e.g., thermal, 1 MeV or 14 MeV) also has an impact on the values of the parameters.

We propose, in analogy of k_0-factors, to introduce a "k_f-factor" for thresholds reactions related to the 411 keV gamma line of gold, Au:

$$k_{f,a,i} = \left(\frac{\overline{\sigma_{f,a,i}}\gamma_a\theta_a}{M_a}\right)/\left(\frac{\sigma_{0,Au}\gamma_{Au}\theta_{Au}}{M_{Au}}\right) \tag{6}$$

with γ as gamma yield probability, θ isotope abundancy and M as the molecular mass. Since $\overline{\sigma_{f,a,i}}$ depends on the shape of the fast neutron spectrum in the irradiation facility, so will $k_{f,a,i}$. By comparing values for $k_{f,a,i}$'s as measured in different irradiation facilities i, cross-facility applicability can be assessed. We do expect to see variation in $k_{f,a,i}$ values across facilities: when the shape of the cross section curve of the reaction of interest differs from that of the fast monitor reaction, the fast neutron spectrum shape will have an impact.

For example, the cross-section behaviour of the reaction Ti-47(n, p) Sc-47 resembles that of Ni-58(n, p) Co-58, see Fig. 1, so the rates for both reactions are expected to be influenced in the same way by the shape of the neutron energy spectrum. For Ti-48(n, p) Sc-48 this is not the case. The reaction has higher cross-sections at higher neutron energies and, as a result, the spectrum-averaged value is more influenced by the shape of the distribution on the high-energy side.

To take the impact of varying fast neutron energy distributions and other possible errors in the k_f's into account, we propose to establish a reference set $k_{f,a,r}$ of $k_{f,a}$ values measured in a specific reference facility. The effect of the shape of the neutron energy spectrum on the effective cross-sections for the fast flux determination (for instance Ni-58(n, p) Co-58) and analysis is corrected for by a newly defined factor $h_{a,i}$ that relates the $k_{f,a,i}$-values applicable to irradiation facility i to the $k_{f,a,r}$-values as follows

$$h_{a,Ni,i} = (k_{f,a,i}/k_{f,a,r})/(k_{f,Ni,i}/k_{f,Ni,r}) \tag{7}$$

To standardize, the fast flux should be monitored by a standard reaction. There are several options. A comparator monitor based on gold/aluminium is often used and Al-27(n, α) Na-24 is therefore an obvious option. Frans De Corte [1] proposed Zr-90(n, 2n) Zr-89 since pure zirconium is also often used for flux calibration. Ni-58(n, p) Co-58 is often chosen because of the low threshold energy and the rather high cross section.

For the determination of the fast flux ratio f_f, we propose to use Ni-58(n, p) Co-58 as monitor with a fixed reference value for $k_{f,Ni,r} = 0.00255$ for the 810.8 keV line, half-life = 70.916 d.

The k_0-formula for analytical use of threshold reactions becomes:

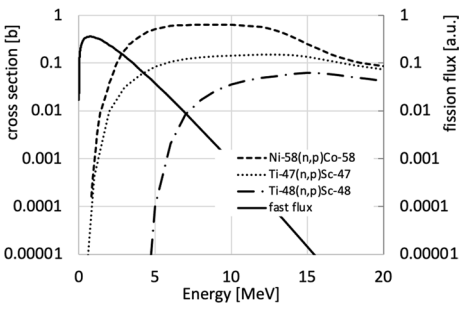

Fig. 1 Cross-section distribution as function of neutron energy

$$\rho_a = \frac{\left(\frac{N_p/t_c}{SDCW}\right)_a}{\left(\frac{N_p/t_c}{SDCw}\right)_m} \cdot \frac{k_{0,m}}{k_{f,a,r}} \cdot \frac{G_{th,m}f + G_{epi,m}Q_{0,m}(\alpha)}{f_f h_{a,i}} \cdot \frac{\varepsilon_{p,m}}{\varepsilon_{p,a}} \quad (8)$$

with

$$f_f = \frac{\Phi_f}{\Phi_{epi}}$$

With ρ as the mass fraction of the analyte to be determined, $\frac{N_p/t_c}{SDCW}$ as the decay corrected specific countrates, ε as gamma full energy efficiency, G as neutron absorption correction factor and $Q_{0,m}(\alpha)$ as activation factor for epi-thermal neutrons all as defined in [1]. Fast neutron absorption in the sample is assumed to be negligible. For analytical use of nuclides produced by threshold reactions this factor, $h_{a,i}$, could be measured or, alternatively, calculated from earlier measured calibration factors. After determination of $h_{a,i}$-values in different irradiation facilities for different reactions, results can be compared and perhaps a relation or model can be found for $h_{a,i}$ to minimize the calibration efforts needed to achieve a desired accuracy. The correction factors for reaction interference are derived from this formula in the "Appendix".

In this paper, we make a first attempt, using experimental work done in the past by Verheijke as well as experimental data obtained at RID.

In his Ph.D. thesis [2], Verheijke describes the use of fission neutrons for the analysis of Ti and Ni in Si-wafers. He measured fast neutrons activation factors (*H)* related to the thermal flux for four reactors (BR1, BR2, HFR and FRJ2) and several tens of irradiation facilities. The irradiations were performed in twofold with Zr, and Au or a Co monitor. *f* was taken from earlier measurements in the used facilities and also provided.

Verheijke found an empirical fast flux model that predicted the observed activation rates for the two water-moderated reactors. He assumed that the fast flux is related to the epi-thermal and thermal flux according to $\Phi_f = c\ \Phi_{th} + e\ \Phi_{epi}$ and determined effective cross sections, were *c* and *e* are constants for specific reactions. He found the effective cross sections to be inversely proportional to $f\,(\Phi_{th}/\Phi_{epi})$ for multiple reactors and channels with *f* ranging from 14 to 100. The data did not yield a unique function for all reactors, only the BR2 and HFR functions matched. The BR1 and FRJ2 data did not match.

Experimental

In order to make a comparison between measurements of $h_{a,i}$, we must select a reference set of $k_{f,a,r}$- values. In this work, we use the experimentally determined $k_{f,a,i}$'s from Reactor Institute Delft as such. These values have been determined by measurements of elemental standards over a period of close to 30 years, starting after the conversion of the RID reactor to low-enriched fuel (see Table 1).

Verheijke's measured activation factors (*H*) for Ni-58(n, p) Co-58, Ti-47(n, p) Sc-47 and Ti-48(n, p) Sc-48 were digitized by the authors and converted to Φ_f/Φ_{epi} vs. Φ_{th}/Φ_{epi} (f_f vs *f*) plots for the three reactions. The fast flux ratios were recalculated using the $k_{f,a,r}$-values from RID. The formula used is based on the formula given by Verheijke: $\Phi_f/\Phi_{th} = c\ H$ [2].

Next, the fast flux ratios for Ti-47 and Ti-48 were divided by the fast flux ratios from the Ni-58(n,p)Co-58 measurement, as proposed. These ratios of ratios are in fact $h_{a,i}$-factors as defined in Eq. 7.

To investigate the effect of the fast spectrum temperature (expressed in E_f) on *h*, using the Maxwell distribution and data from ENDF/B VIII.0, we computed $h_{a,i}$-values as function of the neutron energy E_f from 1.25 to 1.4 MeV and normalized them to the value at 1.35 MeV.

Results

The $k_{f,a,r}$-values determined at RID are shown in Tables 1 and 2.

Verheijke's converted data are shown in Fig. 2, and the $h_{a,i}$-values we derived in Fig. 3.

Table 1 $k_{f,a,r}$ reference values for primary interference nuclides as measured at RID

Reaction	Half life	Energy/keV	k_f
Ni-58—> Co-58	70.916d	511	7.64E-04
Ni-58—> Co-58	70.916d	810.79	2.55E-03
Cu-63—> Co-60	5.271y	1173.2	2.03E-05
Cu-63—> Co-60	5.271y	1332.5	2.04E-05
V-51—> Sc-48	1.821d	983.5	1.65E-07
V-51—> Sc-48	1.821d	1037.5	1.61E-07
V-51—> Sc-48	1.821d	1312.1	1.65E-07
Si-30—> Mg-27	9.462 m	843.8	2.92E-07
Si-30—> Mg-27	9.462 m	1014.4	1.14E-07
Al-27—> Mg-27	9.462 m	843.8	2.30E-04
Al-27—> Mg-27	9.462 m	1014.4	8.98E-05
Al-27—> Na-24	14.97 h	1368.6	9.79E-05
Al-27—> Na-24	14.97 h	2754.0	9.78E-05
Fe-56—> Mn-56	2.583 h	846.8	5.47E-05
Fe-56—> Mn-56	2.583 h	1810.8	1.51E-05
Fe-56—> Mn-56	2.583 h	2113.2	7.91E-06
Mg-24—> Na-24	14.97 h	1368.6	1.08E-04
Mg-24—> Na-24	14.97 h	2754.0	1.08E-04
Na-23—> F-20	11.03 s	1633.6	6.86E-05
Si-28—> Al-28	2.24 m	1779.0	4.27E-04
Sn-117—> Sn-117 m	13.61d	158.6	2.57E-04

The effect of the fast spectrum temperature (expressed in E_f) on $h_{a,i}$-values is shown in Fig. 4.

Discussion

Verheijke's assumption that there is a linear relation between the fast flux, thermal to epithermal flux, at least for the BR2 and HFR rectors is clearly shown in Fig. 2.

Figure 3 shows that the reaction rate for Ti-47 related to Ni-58, or rather the $h_{a,i}$-factor, is almost unity for all reactors and channels. This shows that the effect of the shape of neutron spectrum is similar for Ti-47 and Ni-58 and that the predefined $k_{f,i,r}/k_{f,Ni,r}$ ratios is correctly measured at RID.

Figure 3 also shows that the $h_{a,i}$-factor for Ti-48 becomes higher for more thermalized neutron fluxes (the outlying result for the most thermalized facility of BR1 could be due to the very low fast flux).

These observations are in agreement with the assumption that reactions with equal-shaped cross-section curves, such as Ni-58(n, p) Co-58 and Ti-47(n, p) Sc-47, will all respond equally to changes in fast spectrum energy distribution, but other reactions will require the introduction of an additional parameter such as $h_{a,i}$. One might compare to 1/v and non-1/v reactions in the thermal range, where the non-1/v reactions require the introduction of Westcott factor g(T) to accommodate spectrum temperature differences.

The $h_{a,i}$ for Ni-58(n, p) Co-58 and Ti-47(n, p) Sc-47 show the same unity value over the whole range, but Ti-48(n, p) Sc-48 does not. The shape of the curve in Fig. 3b, seeing Fig. 4, cannot be explained by a decrease of spectrum

Table 2 $k_{f,a,r}$ reference values for analytical useful nuclides as measured at RID

Reaction	Half life	Energy/keV	k_f
Fe-54—> Mn-54	312.2d	834.8	1.74E-04
Pb-204—> Pb-203	2.169d	279.2	1.42E-07
Pb-204—> Pb-204 m	1.117 h	374.7	1.44E-06
Pb-204—> Pb-204 m	1.117 h	899.2	1.61E-06
Pb-204—> Pb-204 m	1.117 h	911.6	1.52E-06
Ti-46—> Sc-46	83.83d	889.3	4.96E-05
Ti-46—> Sc-46	83.83d	1120.5	4.96E-05
Ti-47—> Sc-47	3.341d	159.4	4.17E-05
Ti-48—> Sc-48	1.821d	983.5	1.41E-05
Ti-48—> Sc-48	1.821d	1037.5	1.37E-05
Ti-48—> Sc-48	1.821d	1312.1	1.41E-05
As-75—> As-74	17.78d	511.0	4.31E-06
As-75—> As-74	17.78d	595.9	4.88E-06
As-75—> As-74	17.78d	634.8	1.25E-06
Mo-92—> Nb-92 m	10.15d	934.5	2.13E-05
Mo-95—> Nb-95	34.97d	765.8	5.60E-07
Zr-90—> Zr-89	3.268d	511.0	1.19E-06
F-19—> O-19	26.91 s	197.1	1.01E-04
F-19—> O-19	26.91 s	1356.9	5.86E-05
Si-29—> Al-29	6.56 m	1273.4	9.95E-06
Y-89—> Y-89 m	16.1 s	909.2	3.96E-03
Zn-66—> Cu-66	5.1 m	1039.4	8.26E-07
Ni-58—> Co-58	70.916d	511	7.64E-04
Ni-58—> Co-58	70.916d	810.79	2.55E-03

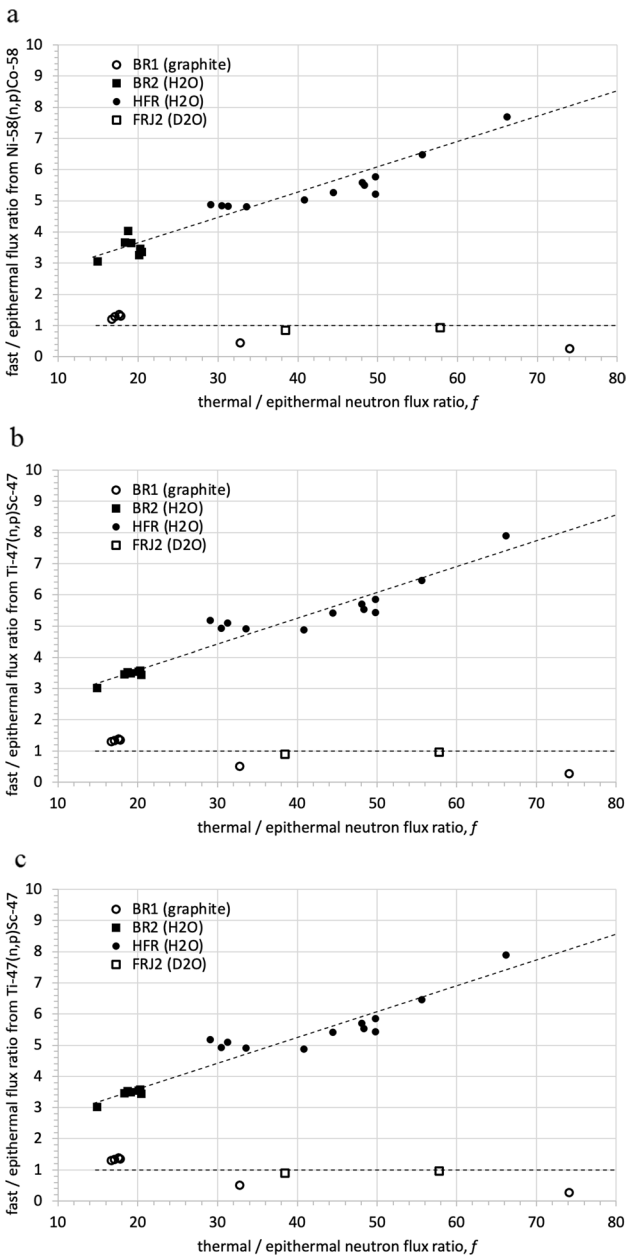

Fig. 2 Fast vs. epithermal neutron flux ratios measured using **a** Ni-58(n, p) Co-58, **b** Ti-47(n, p) Sc-47 and **c** Ti-48(n, p) Sc-48), all recalculated from Verheijke's data [2], based on the newly proposed k_f's (with lines to guide the eye)

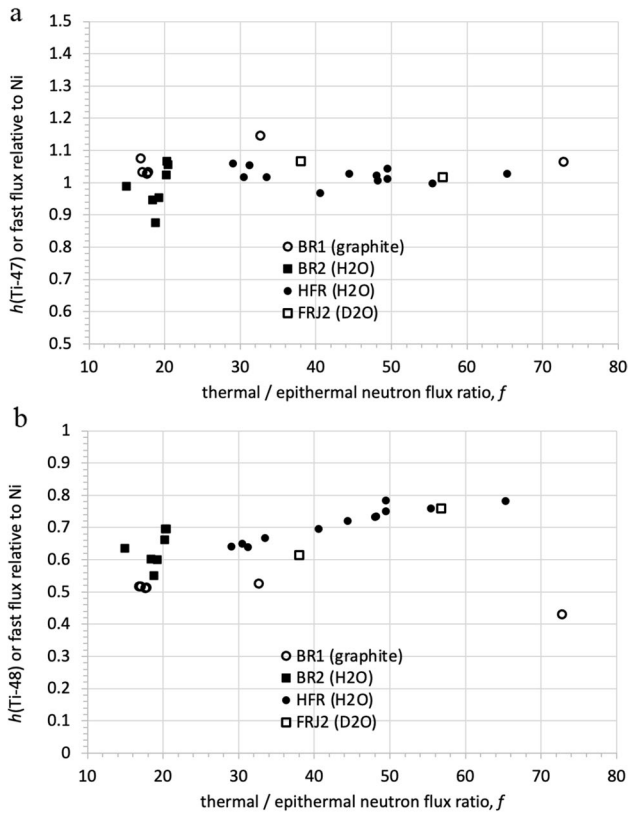

Fig. 3 **a** and **b** $h_{a,i}$-factor for Ti-47(n, p) Sc-47 and Ti-48(n, p) Sc-48

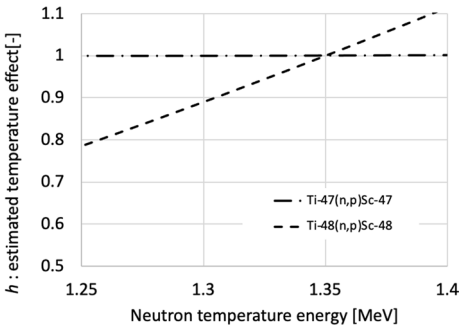

Fig. 4 Effect of changing temperature/energy on the $h_{a,i}$-factors for Ti-47 and Ti-48 as calculated from theory and ENDF/B VIII.0 data

temperature at higher f values, and/or larger distance from the reactor core, so there must be a different reason.

For the Ti-48(n, p) Sc-48 reaction, a $h_{a,i}$-factor can be estimated imprecisely, if f is known for the irradiation facility of interest, by interpolation in Fig. 3b.

Looking at cross sections (in Fig. 5) and theoretical h-values (in Fig. 6) for other relevant reactions, one might conclude, that there are more reactions that are likely to have the same $h_{a,i}$ factors and similar curves in a $h_{a,i}$-f plot for different reactors and channels.

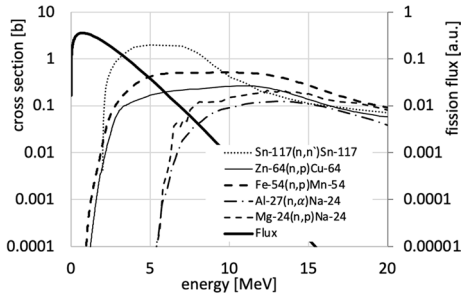

Fig. 5 Activation cross-sections for some useful threshold reactions

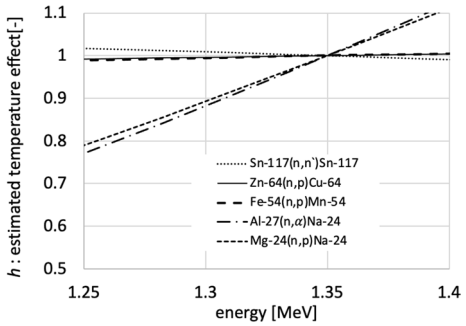

Fig. 6 Effect of changing temperature/energy on the h-factor for some useful threshold reactions

Conclusions

Activation rates in threshold reactions can be taken into account, in NAA, by using the k_f-factors we propose. If a reaction like Ni-58(n, p) Co-58 is used as the standard monitor reaction, the approach will be inaccurate for reactions with cross-section curves with different shapes. To reduce this inaccuracy, reaction and irradiation-facility dependent $h_{a,i}$ factors must be used in addition. These factors can be estimated from theoretical or experimental neutron spectrum data together with known energy-dependent cross-section data. Alternatively, it may be possible to use the correlation we observed between $h_{a,i}$-values and thermal-to-epithermal ratio f.

Appendix 1: Interference corrections factors based on k_f's

Interference reactions can be split in two types:

1. (n, p), (n, α) reactions on target elements (b) that result in a product that also is formed by an (n, γ)-reaction on element (a).

2. (n, n'), (n, 2n) reactions where the target nuclide is one of the natural occurring nuclides of the element under activation and the product is the same as formed by the (n,γ)-reaction.

Correction for threshold reaction interferences of type 1, when the concentration of the interfering element, c_b, is known becomes:

$$c_a = c'_a - I_{bona}.c_b \tag{9}$$

With correction factor for interference:

$$\text{on a (n, }\gamma\text{) reaction: } I_{bona}(g/g) = \frac{k_{f,b}}{k_{0,a}} \cdot \frac{f_{f,b}h_{b,i}}{G_{th,a}f + G_{epi,a}Q_{0,a}(\alpha)} \tag{10}$$

$$\text{on a threshold reaction} : I_{bona}(g/g) = \frac{k_{f,b}}{k_{f,a}} \cdot \frac{h_{b,i}}{h_{a,i}} \tag{11}$$

Correction for threshold reaction interferences of type 2, when the interfering element is the analyte itself and thus unknown, becomes:

$$c_a = c'_a * I_a \tag{12}$$

With correction factor for all interfering reactions, *j*:

$$I_a(-) = \frac{G_{th,a}f + G_{epi,a}Q_{0,a}(\alpha)}{G_{th,a}f + G_{epi,a}Q_{0,a}(\alpha) + \frac{f_f}{k_{0,a}}\sum k_{f,j}h_{j,i}} \tag{13}$$

The type 2 reaction that can interfere the standard k_0 (n, γ)-reaction most is (n, n'), however (n, 2n) can also be of importance. The most important produced isotopes are Se-77 m, Sr-87 m, Cd-111 m, Sn-117 m, Ba-135 m and Ba-137 m, of which Sn-117 m is causing a serious interference up 50% and more. The proposed k_f's for most interfering reactions are given in Table 1.

Declarations

Conflict of interest This paper does not incur potential conflicts of interest. No human test subjects or animals played a part.

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