SHORT COMMUNICATION

Journal of Radioanalytical and Nuclear Chemistry, Vol. 266, No. 1 (2005) 153–157

Epithermal neutron flux characterization of the IEA-R1 research reactor, Sao Paulo, Brazil

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(Received January 25, 2005)

The nonideality of the epithermal neutron flux distribution at a reactor site parameter (α) and the thermal-to-epithermal neutron ratio (f) were determined in three typical irradiation positions of the IEA-R1 reactor of IPEN-CNEN/SP, Sao Paulo, Brazil, using the "Cd-ratio for multimonitor" and "bare bi-isotopic monitor" methods, respectively. This characterization is to be used in the k_0-method of NAA, recently introduced at the IPEN.

Introduction

When applying the k_0-method of instrumental (n,γ) activation analysis with reactor neutrons (INAA), it is found that the general accepted $1/E_n$-epithermal neutron flux distribution is often unacceptable from the standpoint of analysis accuracy.[1] The use of a semiempirical representation[2,3] is better:

$$\varphi_e'(E_n) \sim 1/E^{1+\alpha}$$

which was proved to be satisfactory for INAA needs[4] where α is a measure of the epithermal flux deviation from the ideal, and is a characteristic of the reactor irradiation position. Both positive and negative α-values are reported in the literature,[5] corresponding to a "softened" and a "hardened" epithermal spectrum, as compared to the ideal one. This is illustrated in Fig. 1.

The $1/E^{1+\alpha}$ representation enables easy correction of the resonance integral (and hence of the analytical result) for the deviating epithermal spectrum.[4] Thus, when calculating the concentration of an element in a sample, α should be known to preserve the accuracy of the analysis.

With the same aim, the thermal (subcadmium)-to-epithermal ratio:

$$f = \Phi_{th}/\Phi_e$$

must be well known.[6] The designation "subcadmium" refers to the definition of Φ_{th}, the conventional thermal fluence rate, as:

$$\Phi_{th} = n_s v_0$$

where v_0 is the 2200 m·s^{-1} neutron velocity and n_s is the "subcadmium" neutron density up to 0.55 eV neutron energy. On the other hand, Φ_e, the conventional epithermal neutron fluence rate, is defined as the true epithermal neutron fluence rate per unit ln$E$ interval. The use of f is strictly associated with Q_0 (the resonance integral to 2200 m·s^{-1} cross section ratio), both parameters being linked to the HØGDAHL convention,[7] on which the application of the k_0-method is based.

Experimental and results

All through this work we have investigated α and f in the three characteristic irradiation channels of the IEA-R1 reactor (Fig. 2), i.e., in the "pneumatic transfer tube channel" (Station 4) and in the positions EIRA-24B and EIRA-36B (fixed systems for large irradiation).

We used thin high pure foils as α-monitors considering the self-shielding effects (0.125 mm Zr, 0.125 mm Au, 0.05 mm Co). Relevant nuclear data for the nuclides chosen as monitors are given in Table 1. So as to obtain good statistics on A_{sp} values, the irradiation was repeated 3 times. In all studied positions, the monitors were irradiated during 30 minutes. After appropriate cooling times (24–48 hours), γ-activities were measured on a HPGe (Canberra GMX20190, 1.9 keV for 1332 keV line of ^{60}Co). All monitor spectra were processed using the VISPECT system, developed at IPEN.

The epithermal spectrum shape factor α was obtained[8] as the slope ($-\alpha$) of the straight line when plotting log(T_i) versus log($E_{r,i}$/1 eV), which comes to the same as solving the implicit equation (1 eV omitted) (Fig. 3):

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$$\alpha + \frac{\sum_{i}^{N}\left\{\left[\log(E_{r,i}) - \frac{\sum_{i}^{N}\log(E_{r,i})}{N}\right]\left[\log(T_i) - \frac{\sum_{1}^{N}\log(T_i)}{N}\right]\right\}}{\sum_{1}^{N}\left[\log(E_{r,i}) - \frac{\sum_{1}^{N}\log(E_{r,i})}{N}\right]^2} = 0$$

where N is the number of coirradiated α-monitors, $E_{r,i}$ is the average resonance energy of the monitor i and T_i was given by the "Cd-covered multi-monitor method"[12] as:

$$T_i = \frac{(E_{r,i})^{-\alpha}(A_{sp,i})_{Cd}}{k_{0,Au}(i)\varepsilon_{p,i}F_{Cd,i}Q_{0,i}(\alpha)G_{e,i}}$$

with

$$A_{sp} = (N_p/t_m)/\mathrm{SDCw}$$

where N_p is the measured net peak area, t_m is the counting time, S is the saturation factor, D is the decay factor, C is the counting factor and w is the sample mass (in grams), $k_{0,Au}$ is the tabulated k_0-factors, $\varepsilon_{p,i}$ is the detector efficiency for used γ-line, $F_{Cd,i}$ is the Cd-transmission factor and G_e is the correction factors for epithermal neutron self-shielding.

Table 1. Nuclear data for the nuclides chosen as monitors[8,9]

Monitor	E_r, eV	Q_0	G_{th}*	G_{epi}*	F_{Cd}	Gamma-line, keV	Half-life
$^{197}Au(n,\gamma)^{198_m}u$	5.65 ± 0.40	15.7 ± 0.28	0.989	0.755	0.991	411.8	2.695 d
$^{59}Co(n,\gamma)^{60}Co$	136 ± 7	1.990 ± 0.054	0.986	0.950	1	1173.0	5.271 y
$^{96}Zr(n,\gamma)^{97}Zr$	338 ± 7	248.0 ± 0.4	1	0.966	1	743.6	16.74 h
$^{94}Zr(n,\gamma)^{95}Zr$	6260 ± 250	5.05 ± 0.10	1	0.987	1	(724.2 + 756.7)	64.033 d

* Calculated by program CCOMP,[10] using the DE CORTE methodology[11] and considering the Doppler effect.

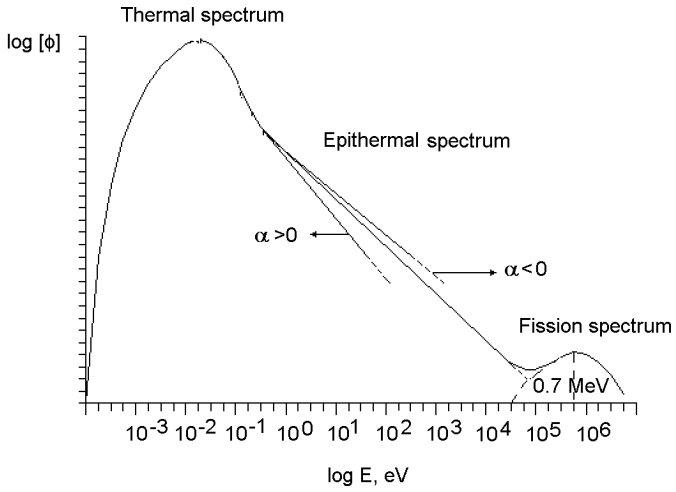

Fig. 1. Epithermal spectrum nonideality

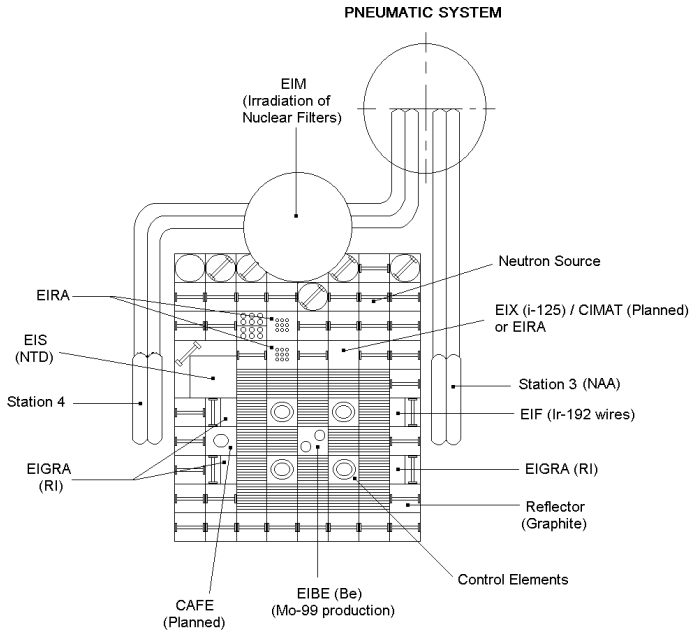

Fig. 2. Ground plan of the IEA-R1 reactor

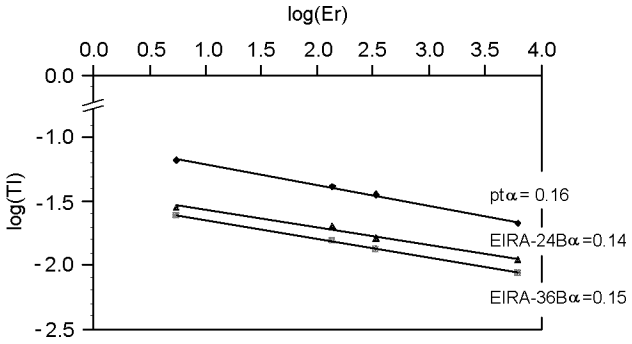

Fig. 3. Experimental α-determination in the three characteristic irradiation positions of the IEA-R1 reactor at IPEN, Brazil

The thermal-to-epithermal neutron flux ratio (f) was determined by the "bare bi-isotopic monitor" method using Zr[13] as:

$$f = \frac{G_{e,1}\dfrac{k_{0,Au}(1)\varepsilon_{p,1}}{k_{0,Au}(2)\varepsilon_{p,1}}Q_{0,1}(\alpha) - G_{e,2}\dfrac{A_{sp,1}}{A_{sp,2}}Q_{0,2}(\alpha)}{G_{th,2}\dfrac{A_{sp,1}}{A_{sp,2}} - G_{th,1}\dfrac{k_{0,Au}(1)\varepsilon_{p,1}}{k_{0,Au}(2)\varepsilon_{p,1}}}$$

where indexes of $1 = {}^{97}\mathrm{Zr}/{}^{97m}\mathrm{Nb}$ (743 keV), $2 = {}^{95}\mathrm{Zr}$ (724.2+756.7 keV), G_{th} is the correction factor for thermal neutron self-shielding and $\varepsilon_{p,1} = \varepsilon_{p,2}$. Due to the single-decayed gamma-lines, it is allowed to position the Zr monitor as close as possible to the detector cap.

Table 2 shows the results for α and f. Average values and all statistical processing was performed using the BABXEL system.[14] The quoted uncertainties were calculated according to the error propagation study given in Reference 12. Though, relatively high (~10–30%), inherent α-uncertainties are satisfactory for NAA needs, due to the large error reduction factor when calculating the concentration in the absolute or a single comparator method.[4] In this regard, large relative uncertainties are acceptable for lower absolute α's.

Positive α-values correspond to a "softened" (strongly thermalized) epithermal spectrum, which is indeed to be expected due to the reactor configuration and to ^{235}U enrichment (<20%) in the fuel elements.[15] The practically non-variation of α-values in all studied position is relevant. The relative low f-value in the pneumatic channel is an evidence of the presence of a great portion of epithermal neutrons in that position.

To evaluate the appropriate results obtained in the reactor characterization for k_0-INAA, five replicas of the certified reference material SL-1 Lake Sediment[16] were irradiated in the EIRA-36B irradiation position. The concentrations determined by the HØGDALH convention[11] for "long lived" elements (t_i = 8 h, t_d = 15 d and t_m = 1.0–2.5 h) and their random errors are presented in Table 3. The satisfaction of the precision tests installed in the BABXEL system is also showed.

A very good coincidence was observed between the determined average concentrations and the reported ones (Fig. 4). Comparing the results for the SL-1 certified elements, several concentrations were determined with a better accuracy than reported. On the other hand, the application of the different precision tests for all determined concentrations showed positive results. A good accuracy and precision are important evidence that the main reactor neutron flux parameters for k_0-INAA were well determined.

Table 2. α and *f*-values determined in the irradiation positions of the IEA-R1M reactor

Channel	α (Cd-ratio)	*f* (bare)
PT	0.16 ± 0.01	11.1 ± 0.6
EIRA-24B	0.14 ± 0.01	26.5 ± 0.2
EIRA-36B	0.15 ± 0.01	31.2 ± 0.3

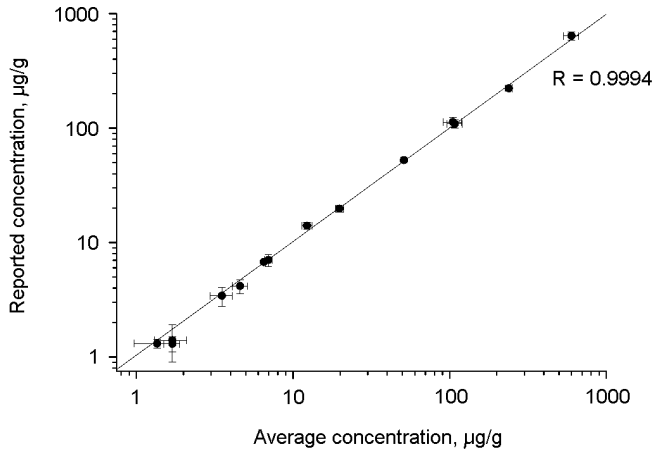

Fig. 4. Comparison between the reported and determined concentrations in CRM-IAEA-SL-1

Table 3. k_0-INAA of the certified reference material IAEA-SL-1 "Lake Sediment"

Element	Average concentration, µg/g	Reported concentration, µg/g
Lu	0.52 ± 0.07	0.54*
Hf	4.6 ± 0.5	4.16 ± 0.58
Mo	1.7 ± 0.2	1.3*
Fe (%)	6.52 ± 0.13	6.74 ± 0.17
Yb	3.52 ± 0.56	3.42 ± 0.64
Ba	600 ± 66	639 ± 53
Th	12.3 ± 0.9	14 ± 1
Tb	1.7 ± 0.4	1.4*
Cr	108 ± 12	109 ± 9
Br	[+]	6.82 ± 1.73
Sb	1.36 ± 0.39	1.31 ± 0.12
Cs	6.97 ± 0.39	7.01 ± 0.88
La	51.1 ± 1.0	52.6 ± 3.1
Rb	105 ± 14	113 ± 11
Zn	239 ± 12	223 ± 10
Co	19.8 ± 1.2	19.8 ± 1.5
Ca (%)	[+]	0.25*

* Non-certified element in CRM SL-1.

[+] Gamma-lines were not observed in the SL-1 spectra.

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The present work was performed under IAEA Research Project BRA/0/017-32.

The authors are grateful to the International Atomic Energy Agency for the partial financial support of this research. Gratitude is also to the IAE-R1M reactor group for its help during the experiments.

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