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Original Article

Re-characterization of pneumatic station position for short irradiations of IEA-R1 reactor for use in k_0-method

Flores[a*], J.P.O.; Semmler[a], R.; Dias, M.S.; Koskinas[a], M.F.; Moreira[a], D.S.; Yamazaki[a], I.M.; Silva[a], P.S.C. and Maihara,V.A.

[a] Instituto de Pesquisas Energéticas e Nucleares (IPEN/CNEN), Av. Professor Lineu Prestes 2242 05508-000, Sao Paulo, Brazil.

*Correspondence: joaooflo@gmail.com

Abstract: This work presents the re-characterization of pneumatic station irradiation position of IEA-R1 reactor for use in k_0-INAA method. The thermal-epithermal neutron flux ratio f and the epithermal neutron flux distribution factor α of the IPEN IEA-R1 reactor were determined at the pneumatic irradiation station. To obtain these values, the "bare triple monitor" method was used with ^{198}Au –^{97}Zr – ^{95}Zr. To evaluate its reproducibility, the temporal variation of these parameters was analyzed, with validation carried out through irradiation of reference materials of geological origin G-S-N (granite) and biological INCT-MPH-2 (mix of herbs). The results obtained for f and α show that such values are reproducible and compatible with each other, according to the normal Gaussian distribution for up to 1 standard deviation of confidence. The results obtained for the elements detected in the samples agree with the recommended values, with relative errors (bias) less than 10%, except in the case of Mn in G-S-N (16.3%). These results indicate that it is possible to routinely use this method for LAN-IPEN researchers to analyze geological and biological samples.

Keywords: k_0-INAA, neutron activation method, neutron flux.

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Re-caracterização da posição da estação pneumática para irradiações curtas do reator IEA-R1 para uso no método k_0

Resumo: Este trabalho apresenta a re-caracterização da posição de irradiação da estação pneumática do reator IEA-R1 para utilização no método k_0-INAA. A razão do fluxo de nêutrons térmicos-epitérmicos e o fator da distribuição de fluxo de nêutrons epitérmicos do reator IEA-R1 do IPEN foram determinados na estação pneumática de irradiação. Para obter esses valores, foi utilizado o método "bare triple monitor" com ^{198}Au–^{97}Zr–^{95}Zr. Para avaliar sua reprodutibilidade, foi analisada a variação temporal desses parâmetros, sendo a validação efetuada por meio de irradiações de materiais de referência de origem geológica G-S-N (granito) e biológica INCT-MPH-2 (mix de ervas). Os resultados obtidos para α e f mostram que tais valores são reprodutíveis e são compatíveis entre si, segundo a distribuição normal gaussiana para até 1 desvio padrão de confiança. Os resultados obtidos para os elementos detectados nas amostras concordam com os valores recomendados, com erros relativos (bias) menores que 10%, exceto no caso do Mn no G-S-N(16,3%). Estes resultados indicam que é possível viabilizar o uso desse método de forma rotineira para os pesquisadores do LAN-IPEN, para análise de amostras geológicas e biológicas.

Palavras-chave: k_0-INAA, método de ativação neutrônica, fluxo de nêutrons.

1. INTRODUCTION

The k_0-method[1] is a quasi-absolute neutron activation analysis technique for multielement determination. Due to its excellent accuracy and other advantages is a powerful option to the comparative method, which has been successfully applied in numerous studies carried out at the IPEN Neutron Activation Analysis Laboratory (LAN)[2].

To apply the k_0-method, it is necessary to perform a precise characterization of the irradiation position in the nuclear reactor, determining the spectral parameters f (the ratio between the thermal and epithermal neutron fluxes) [3], and α (which describes the deviation of the epithermal neutron flux from the ideal $1/E$ energy dependence)[3,4]. These parameters are intrinsic to each irradiation position and reactor configuration and are essential for the correct application of the k_0 formalism. In addition, the characterization of the counting geometry requires a precise determination of the detector efficiency curve, which relates the absolute full-energy peak efficiency to the gamma-ray energy for a fixed and well-defined source-to-detector geometry.

In this work, the detector efficiency was determined by efficiency calibration using calibration sources, following the recommendations of De Corte[3]. The characterization of the counting geometry refers to the strict control and reproducibility of the source-to-detector distance, sample positioning, and counting conditions, ensuring that the efficiency curve can be reliably applied to all measured samples and monitors. This procedure is required to minimize systematic uncertainties in activity determination and, consequently, in elemental concentration calculations[3].

The k_0-INAA equation used in this work can be written, in simplified form, as:

$$\rho_a = \frac{A_{sp,a}}{A_{sp,Au}} \cdot \frac{\varepsilon_{p,Au}}{\varepsilon_{p,a}} \cdot \frac{1}{k_{0,Au}(a)} \cdot \frac{G_{th,Au} \cdot f + G_{e,Au} \cdot Q_{0,Au}(\alpha)}{G_{th,a} \cdot f + G_{e,a} \cdot Q_{0,a}(\alpha)} \qquad (1)$$

where:

a – analyte;

α– measure for the epithermal neutron fluence rate distribution;

A_{sp} – specific counting rate;

ε – detection efficiency of the gamma-ray peak at energy E_γ;

$k_{0,Au}(a)$ – k_0 factor of the analyzed isotope, referenced to the gold monitor;

$G_{th,}$ – self-shielding correction factor for thermal neutrons;

G_e – self-shielding correction factor for epithermal neutrons;

Q_0 – ratio between the resonance integral and the thermal neutron cross section.

The objectives of the present work were to re-determine the parameters α and f, in the irradiation channel of the pneumatic station of the IEA-R1 reactor using the bare triple monitor method and the bi-monitors [4], respectively. These methods are essentials for in-situ α and f parameters determination in NAA[3]. In the bare triple method a set of monitors (^{94}Zr, ^{96}Zr and ^{197}Au) is irradiated, without cadmium cover, in the same conditions as the sample and subsequently measured on the detector. This method is of great importance for routine flux monitoring. For f determination, where the bi-monitor was applied, only the ^{94}Zr and ^{96}Zr monitors are required[3]. The monitors and the relevant nuclear data that were used in this work as shown in table 1.

Table 1 : Some parameters of the reactions used in the Au-Zr bare triple-monitor method[8].

Monitor	Q_0	Half-life	$k_{0,Au}$
$^{197}Au(n,\gamma)^{198}Au$	15.71	2.695 (d)	1
$^{94}Zr(n,\gamma)^{95}Zr$	5.31	64.02 (d)	2.00E-04
$^{96}Zr(n,\gamma)^{97}Zr$	251.6	16.74 (h)	1.24E-05

To evaluate the accuracy of the results, bias (%) and U-score number test were applied to the results obtained in the analysis of the reference materials granite G-S-N and INCT-MPH-2 mixed polish herbs.

2. MATERIALS AND METHODS

The efficiency curve for the HPGe detector was determined at a "reference" position, at 100 mm source to detector distance, using calibration sources of ^{133}Ba, ^{60}Co, ^{137}Cs, ^{152}Eu, ^{22}Na and ^{166m}Ho, with energies ranging from 121 keV to 1408 keV [5]. All the measurements were done at the reference position.

The parameters α and f were determined irradiating a set consisting of ~40 mg of a Zr foil in square shape, (Aldrich Chemical Company, 0.25 mm thick, purity 99.8 %) together with ~8 mg of Al-0.1 %Au wire (Certified Reference Material IRMM-530R) and ~16mg of a Ni foil in square shape, (Aldrich Chemical Company, 0.125 mm tick, purity 99.9%) was used in fast neutron flux determination and was irradiated together with Au and Zr monitors. The fast neutron flux was determined only once, using the ^{58}Ni(n,p)^{58}Co reaction with the follow nuclear reaction data, energy of 810.76 keV with 99.4% of gamma probability and cross section of 1.07 $\times 10^{-25}$ cm^2, used for the calculation of the fast neutron flux.

The thermal and the fast neutron fluxes were determined by the equations 2 and 3 respectively[3].

$$\phi_{th} = \frac{f \cdot A_{sp} \cdot 3.47 \cdot 10^6}{(f + Q_0(\alpha)) \cdot \varepsilon_p} \qquad (2)$$

$$\phi_f = \frac{M \cdot A_{sp}}{N_a \cdot \theta \cdot I_\gamma \cdot \varepsilon_p \cdot \sigma} \qquad (3)$$

where:

M – molar mass;

A_{sp} – specific area;

ε_p – nickel efficiency on the reference position;

I_γ – gamma emission probability;

θ – isotopic abundance;

σ – cross section;

N_a - Avogadro's constant;

A total of nine independent irradiations were performed over a period of two years. For each one, individual values of α and f were calculated, allowing the evaluation of temporal reproducibility. The monitors irradiation time was of 2.5 min under a thermal neutron flux of (1.90 ± 0.15) × 10^{12} cm^{-2} s^{-1}.

Aliquots of ~100 mg of the geological reference material G-S-N granite from "Association Nationale de la Recherche Technique (ANRT), Vosges (France), ~88 mg of the biological reference material INCT-MPH-2 mixed polish herbs from the Department of Analytical Chemistry at the Institute of Nuclear Chemistry and Technology in Warsaw, Poland and monitors (~5 mg Al-0.1 %Au alloy) were sealed in polyethylene capsules and irradiated for 30 seconds at the pneumatic station irradiation position of the IEA-R1 reactor, and the induced gamma-activities were measured using the calibrated gamma-spectrometer.

The detector used for acquisition of the gamma ray spectra was a HPGe detector (Canberra model GX3018), coaxial geometry and relative efficiency of 30% and a resolution of 1.8 keV, at the energy of 1332.5 keV of ^{60}Co. The associated electronics is the conventional one for simple spectroscopy. The detector is connected to a Canberra DSA-LX multichannel analyzer integrated into a microcomputer. Gamma ray spectra were collected and processed using the Maestro software

3. RESULTS AND DISCUSSIONS

The efficiency[5] curve for the HPGe spectrometer is represented in Figure 1.

Figure 1: Efficiency curve as a function of energy, obtained for the HPGe detector at 100 mm distance source-detector.

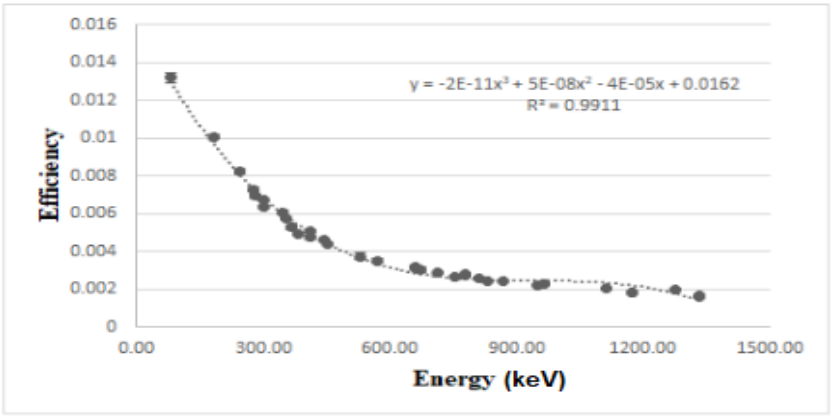


Table 2 : Values of α, f and ϕ_{th} with their respective uncertainties[5].

Irradiation	α	f	ϕ_{th} 10^{12} cm^{-2} s^{-1}
1	0.0395 ± 0.0050	36.1 ± 1.3	1.85 ± 0.15
2	0.0397 ± 0.0092	36.7 ± 1.4	1.91 ± 0.23
3	0.0362 ± 0.0094	37.8 ± 1.6	1.87 ± 0.21
4	0.0378 ± 0.0094	36.8 ± 1.8	1.89 ± 0.18
5	0.0366 ± 0.0089	37.7 ± 1.5	1.93 ± 0.33
6	0.0386 ± 0.0087	35.4 ± 1.5	1.91 ± 0.15
7	0.0395 ± 0.0087	35.3 ± 1.4	1.99 ± 0.31
8	0.0398 ± 0.0090	37.2 ± 1.4	1.91 ± 0.17
9	0.0429 ± 0.0018	38.4 ± 1.7	1.89 ± 0.22

The fast neutron flux value obtained for the irradiation 2 was (2.04 ± 0.25)×10^{11}cm^{-2} s^{-1}.

A comparison was made between the flux parameters with the work done by Puerta[7], for a better evaluation, as shown in table 3, we observed that these parameters are in agreement with the exception of the fast neutron flow, the value obtained for the thermal flux represents the average of the nine irradiations.

Table 3 : Comparison between Flores and Puerta flux parameters.

Parameters	Flores[5]	Puerta[7]
Thermal neutron flux, ϕ_{th} ×10^{12} cm^{-2} s^{-1}	1.90 ± 0.15	1.82 ± 0.05
Fast neutron flux, ϕ_{fast} ×10^{11} cm^{-2} s^{-1}	2.04 ± 0.25	3.66 ± 0.37
Thermal to ephitermal flux ratio, f	36.8 ± 1.5	35.6 ± 1.1
Deviation of the epithermal neutron flux distribution, α	0.0388 ± 0.0097	0.0288 ± 0.0058

To evaluate the results obtained in this work, the concentration values obtained were compared to the values recommended by the certificates using the statistical criteria commonly used in laboratory tests: U-score[9] and relative error (Bias) as shown in table 4.

Analyzing the results in terms of percentage of error in relation to the certified values (relative errors), it is seen that the errors were between 0 and 10%, except for Mn (16.3%). It is important to note that the errors were randomly above and below the recommended values, showing that there are no systematic errors.

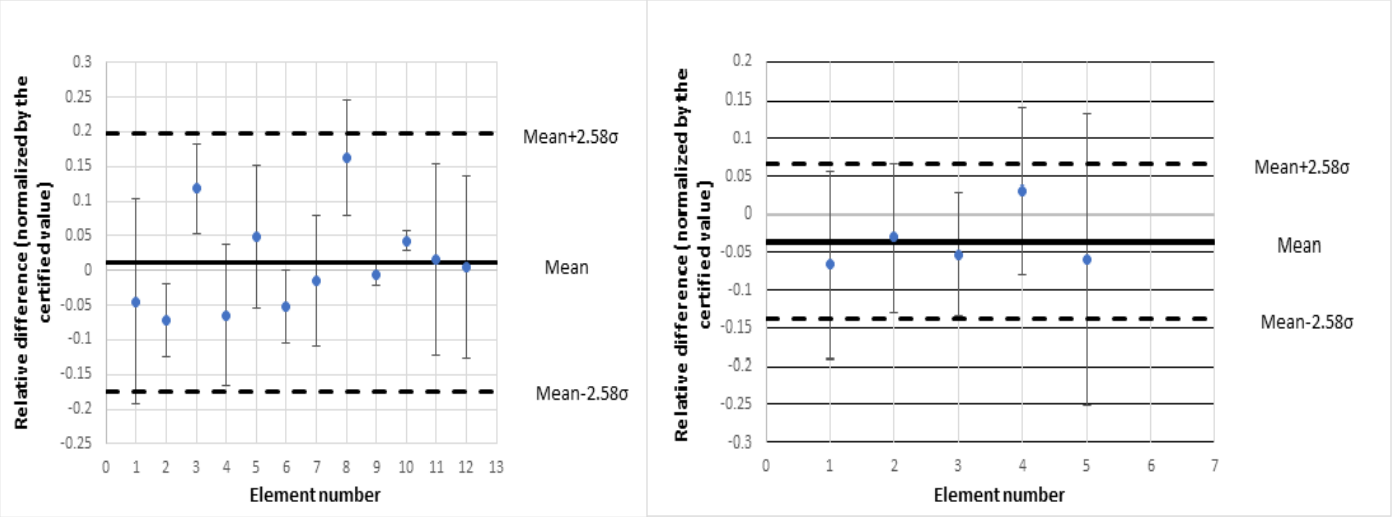
The U-score values showed that all results, with the exception of the value for Na and Mn and Mn in G-S-N and Na for INCT-MPH-2, are within a 95% confidence interval (if 1.96 is considered the limit value for the U-test for a 95% probability interval). This means that the results do not differ significantly from the expected value. The value of Na and Mn can be accepted by this criterion within a 99.5% confidence interval (U-score ≤ 2.58). A visual form of the Z test is through the Bland-Altman plot [10], which is used to analyze the agreement of different assays or analysis methods. This graph shows the difference values between the corresponding measurement pairs (Certificate - Experimental) as a function of the arithmetic mean between the respective measurement values. Figure 2 show the Bland-

Altman plot for the data in Table 4, where the differences between measurements were normalized by the respective mean values, to facilitate visualization. The solid horizontal line represents the arithmetic mean of the differences (Certificate - Experimental) and the dotted lines show the values corresponding to the 99% confidence interval (mean ± 2.58 × Standard deviation of Certificate - Experimental).

Table 4 : Values obtained for the reference material G-S-N and INCT-MPH-2 in short irradiations.

G-S-N				
Element	**Certified values**	**Experimental results**	**Bias(%)**	**U-score**
Al (%)	7.76 ± 0.05	7.81 ± 0.11	0.6	0.41
Ba (μg/g)	1400 ± 44	1500 ± 60	7.1	1.34
Dy (μg/g)	3.1 ± 0.3	3.3 ± 0.1	6.4	0.43
Eu (μg/g)	1.7 ± 0.1	1.5 ± 0.1	5.9	1.39
K (%)	3.84 ± 0.05	3.82 ± 0.2	0.5	0.1
Mg (%)	1.39 ± 0.05	1.41 ± 0.12	1.4	0.31
Mn (μg/g)	430 ± 30	360 ± 19	16.3	1.95
Na (%)	2.8 ± 0.04	2.68 ± 0.01	4.3	2.4
Sr (μg/g)	570 ± 19	600 ± 23	5.3	1.01
Ti (%)	0.41 ± 0.03	0.39 ± 0.03	4.9	0.47
V (μg/g)	65 ± 8	64 ± 4	1.5	0.11
W (μg/g)	450 ± 63	470 ± 21	8.9	0.56
INCT-MPH-2				
Element	**Certificate values**	**Experimental results**	**Bias(%)**	**U-score**
Al (μg/g)	670 ± 111	710 ± 65	6	0.31
Br (μg/g)	7.71 ± 0.61	8.22 ± 0.73	6.6	0.54
K (%)	1.91 ± 0.12	1.85 ± 0.17	3.1	0.29
Mg (%)	0.292 ± 0.018	0.301 ± 0.022	3.1	0.36
Mn (μg/g)	191 ± 12	201 ± 10	5.2	0.64

Figure 2: Bland-Altman plot for 99.5% confidence interval for G-S-N (a) and INCT-MPH-2 (b).



4. CONCLUSIONS

The α and f values were re-determined over a 2-year period and reproducibility was observed in the results. The values obtained in this work are in agreement with those obtained by Oliveira [5] according to the Gaussian normal distribution, for 1 sigma of confidence, as long as the reactor power remains stable at 4.5 MW, the values of α and f have remained compatible. The main focus of the study was to show that it is possible to use the k_0-INAA method without the need to determine the α and f parameters every time the samples are irradiated in the IEA-R1 reactor in the position of the pneumatic station, aiming to make this method a routine in the neutron activation analysis laboratory, requiring the measurement of parameters only when there is a change in the core configuration.

The results obtained showed a good performance of the k_0-INAA method to analyze different geological or biological matrices, providing reliable results for different elements, with varying concentration ranges. This verifies the feasibility of using such a method routinely at LAN-IPEN, thus increasing its analytical potential for geochemical and environmental studies and maintaining data quality.

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DATA AVAILABILITY STATEMENT

The authors declare that the data supporting the results of this study are available in the article. Derived data supporting the conclusions of this study are available upon request from the corresponding author.

REFERENCES

[1] SIMONITS, A.; DE CORTE, F.; HOSTE, J.; Single-Comparator methods in reactor neutron activation analysis. **Journal of Radioanalytical and Nuclear Chemistry**, v. 24, p. 31-46, 1975.

[2] LANGE, C. N.; CAMARGO, I. M. C.; FIGUEIREDO, A. M. G. M.; CASTRO, L.; VASCONCELLOS, M. B. A.; TICIANELLI, R. B. A Brazilian coal fly ash as a potential source of rare earth elements. **Journal of Radioanalytical and Nuclear Chemistry**, v. 311, p. 1235-1241, 2017.

[3] DE CORTE, F. The k_0-Standardization Method: A move to the optimisation of neutron activation analysis, Rijksuniversiteit Gent, Faculteit Van de Wetenschappen, Gent, Belgium, 1986.

[4] DE CORTE, F.; MOENS, L.; SIMONITS, A.; DE WISPERLAERE, A.; HOSTE, J. Instantaneous α-determination without Cd-cover in the $1/E^{1+\alpha}$ epithermal neutron spectrum. **Journal of Radioanalytical and Nuclear Chemistry**, v. 52, p. 295, 1979.

[5] FLORES, J. P. O.; SILVA, P. S. C.; DIAS, M. S.; KOSKINAS, M. F.; MOREIRA, D. S.; YAMAZAKI, I. M. Calibration of the short irradiation facility for k_0 - NAA implementation at the IEA-R1 reactor. **Brazilian Journal of Radiation Sciences**, 9(1A), São Paulo, 2021.

[6] DE CORTE, F.; SIMONITS, A. Recommended nuclear data for use in the k_0 standardization of neutron activation analysis. **At. Data Nucl. Data Tables**, v. 85, p. 47-67, 2003.

[7] PUERTA, C. D. Aplicação do método k_0-INAA no laboratório de análise por ativação com nêutrons do Ipen utilizando o programa k_0-IAEA. Análise de amostras biológicas. 2013. 202 f. Dissertação (Mestrado em Tecnologia Nuclear), Instituto de Pesquisas Energéticas e Nucleares, IPEN-CNEN, São Paulo. Disponível em: ‹http://repositorio.ipen.br/› (data de consulta no formato: 16/02/2024).

[8] Jaćimović R, k0 Database, available in: https://www.kayzero.com/k0naa/k0naaorg/Nuclear_Data_SC/Entries/2020/8/24_Update_of_k0-database_I-128.html. Accessed: 05 May 2022.

[9] IAEA – International Atomic Energy Agency. Proficiency Testing by Interlaboratory Comparison: Analytical Laboratories in Nuclear Institutions. IAEA-TECDOC-1210, Vienna, 2001.

[10] BLAND, J.; ALTMAN M. D. G. Statistical methods for assessing agreement between two methods of clinical measurement. **The Lancet**, v. 327, n. 8476, p. 307–310, 1986.

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