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Re-characterization of pneumatic station irradiation position of IEA-R1 reactor for use in k_0 - INAA method

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1. Introduction

The neutron activation method k_0[1] is a quasi-absolute neutron activation analysis technique which, because of its excellent accuracy and practicality emerged as an alternative and complementary technique to the comparative method, which has been used successfully in numerous experiments carried out by the IPEN Neutron Activation Analysis Laboratory (LAN)[2].

In this k_0 method there is a need for the precise characterization of the irradiation facility and characterization of geometry conting. For the caracterization of irradiation system it is necessary to determine the ratio between the thermal and epithermal neutron fluxes (f) and the parameter (α) related to the distribution of epithermal neutron flux approximately given by $1/E^{1+\alpha}$. These parameters are characteristic of the irradiation position in the nuclear reactor[3]. For the characterization of geometry counting it is required a precise determination of detector efficiency curve.

The objectives of the present work was to re-determine and re-analyse the parameters α and f, checking temporal variation of these parameters in the irradiation channel of the pneumatic station of the IEA-R1 reactor using the bare triple monitor method and the bi-monitors [4]. 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. Methodology

The efficiency curve for the HPGe detector was determined at a "reference" position, at approximately 100 mm source to detector distance, using standard 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].

The parameters α and f were determined by using the "bare triple-monitor" method with ^{197}Au-^{96}Zr–^{94}Zr and irradiating a set consisting of $\sim$ 40 mg of a Zr foil (Aldrich Chemical Company, 0.25 mm thick, purity 99.8 %) together with $\sim$ 8 mg of Al-0.1 %Au wire (Certified Reference Material IRMM-530R).

A set of monitors were irradiated and for each irradiation α and f values were obtained. These irradiations were carried out over 2 years, in total there were 9 irradiations. The monitors irradiation time was of 2.5 min under a thermal neutron flux of $(1.90 \pm 0.15)10^{12} cm^{-2}s^{-1}$. The relevant nuclear data of monitors can be seen in reference [6].

Aliquots of $\sim$ 100 mg of the geological reference material G-S-N granite, $\sim$ 88 mg of the biological reference material INCT-MPH-2 mixed polish herbs and and standards ($\sim$ 5 mg Al- 0.1 %Au alloy) were sealed in polyethylene capsules and irradiated at the pneumatic station irradiation position of the IEA-R1 reactor, and the induced gamma-activities were measured using the calibrated gamma-spectrometer.

3. Results and Discussion

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

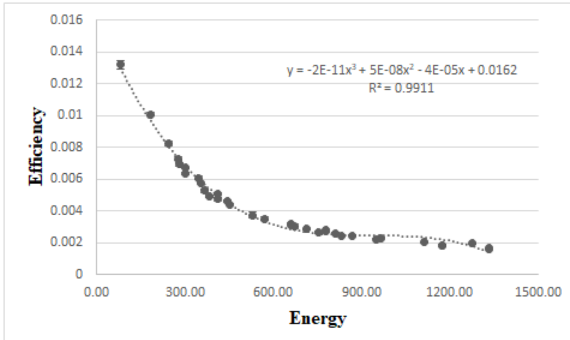

Figure 1: Efficiency curve as a function of energy, obtained for the HPGe detector.

Table I presents the values of the flux parameters (f, α and thermal neutron flux) for the pneumatic station of the IEA-R1 reactor. It is possible observe the reproducibility of the results and the compatibility between the parameters with a confidence level of 99.5%, that is, up to three sigma deviations.

Table I: Values of α, f and ϕ_{th} with their respective uncertainties.

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 was determined using the ^{58}Ni(n,p)^{58}Co reaction. The value obtained was $(2.04 \pm 0.25)10^{11} cm^{-2} s^{-1}$.

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

Table II: Comparison between Flores and Puerta flux parameters.

Parameters	Values	
	Flores[5]	Puerta[7]
Thermal neutron flux, $\phi_{th} 10^{12} cm^{-2} s^{-1}$	1.90 ± 0.15	1.82 ± 0.05
Fast neutron flux, $\phi_f 10^{11} cm^{-2} s^{-1}$	2.04 ± 0.25	3.66 ± 0.37
Thermal to ephitermal flux ratio, f	36.8 ± 1.5	$35.6 \pm 1,1$
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 and relative error (Bias) as shown in table III.

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

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

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 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 $\leq$ 2.58).

A visual form of the Z test is through the Bland-Altman plot, 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 III , 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 $\pm$ 2.58 $\times$ Standard deviation of Certificate - Experimental).

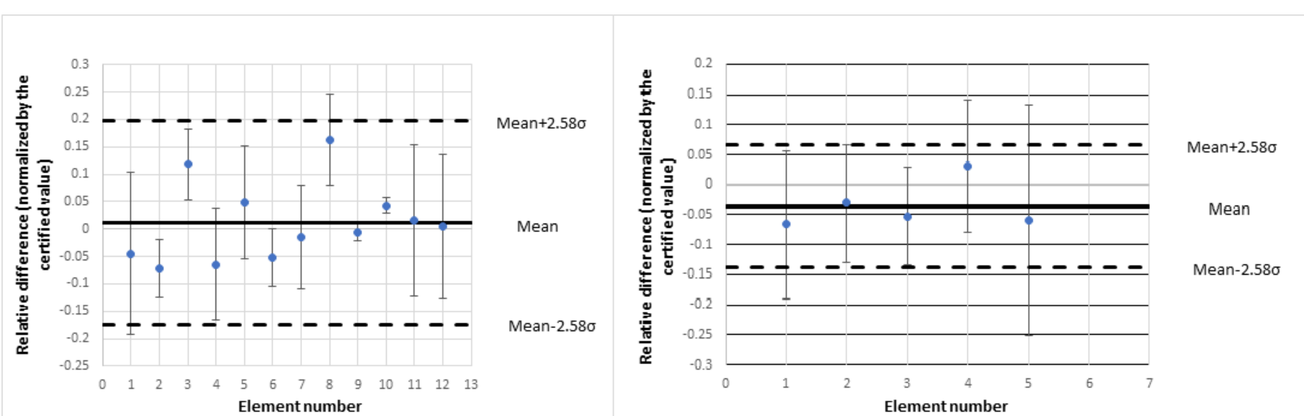

Figure 2: Bland-Altman plot for 99.5% confidence interval for G-S-N (a) and INCT-MPH-2 (b).

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. 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 alpha 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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