



Calibration Of The Short Irradiation Facility For k_0 - NAA Implementation At The IEA-R1 Reactor

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ABSTRACT

The short irradiation facility of the IEA-R1 nuclear research reactor at IPEN, São Paulo, Brazil, has been used for short irradiation of samples for the purpose of determining the concentration of elements of these samples through the use of the instrumental neutron activation analysis technique. With the aim of determine precisely the reactor parameters α and f , for implementing the k_0 -NAA method at the Neutron Activation Analysis Laboratory (LAN), was used the bare triple method. In this method, a set of three neutron flux monitors were

irradiated without Cd-cover. The efficiency curve of the gamma-ray spectrometer used was determined by measuring calibrated radioactive sources at the commonly used counting geometries. The results for the parameters α and f were respectively 0.0384 ± 0.0016 and 35.67 ± 0.26 . This value of f shows that the neutrons in the irradiation position are well thermalized. The variation of these parameters was studied with time and the reproducibility was verified.

Keywords: k_0 method, Neutron Activation Analysis, Neutron flux parameters.

1. INTRODUCTION

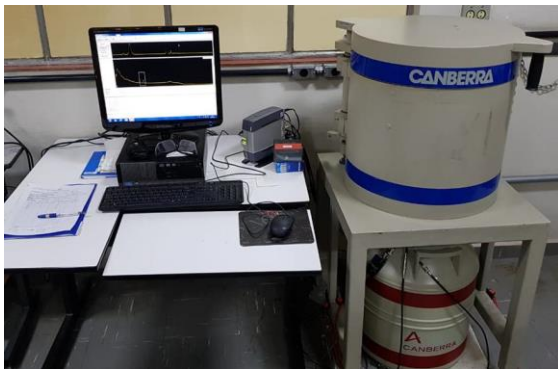
The neutron activation method k_0 [1], developed by the Institute for Nuclear Sciences in Gent, Belgium, is a quasi-absolute neutron activation analysis technique which, because of its excellent accuracy and practicality, emerged as an alternative and complementary technique to the comparative method, which has been used successfully in numerous experiments carried out by the IPEN Neutron Activation Analysis Laboratory (LAN) [2-7], however, is sometimes not feasible using neutron activation analysis in its usual form (comparative method): in other words irradiation and measurement of a standard for each element to be determined. So, the k_0 method presents some advantages over the classical one, as no need for preparation of a large number of standards, provide the same irradiation condition for the sample and the standard and decrease sample analysis time.

In this k_0 method there is a need for the precise characterization of the irradiation facility and characterization of the counting geometry. For the characterization of the 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 important characteristics of the irradiation position in a nuclear reactor [8]. For the characterization of the counting geometry, a precise determination of the detector efficiency curve is required.

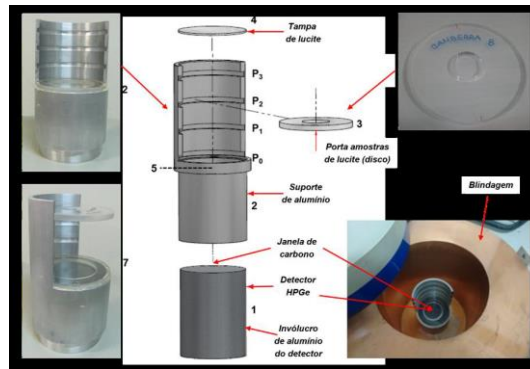
The main objective of the present work was to determine the parameters α and f in the irradiation channel of the pneumatic station of the IEA-R1 reactor by using the bare triple monitor method and the bi-monitors [1]. A precise characterization of this position of irradiation will enable the implementation and use of the k_0 method of neutron activation in the Activation Laboratory for short time irradiations.

2. MATERIALS AND METHODS

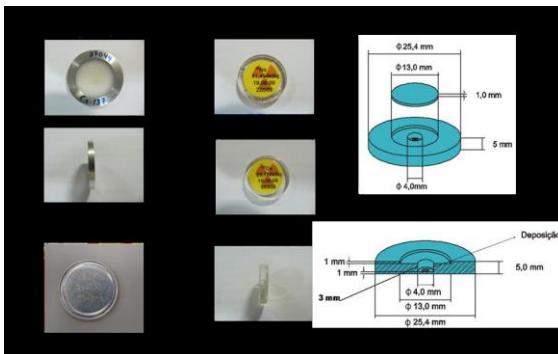
The detector used for acquisition of the gamma ray spectra was a Hiperpure Germanium (HPGe) detector, Canberra model GX3018 (Canberra 8), coaxial geometry and relative efficiency of 30% and a resolution of 1.8 keV, at the energy of 1332.5 keV of ^{60}Co . The associated electronics is the conventional one for simple spectroscopy. The detector is connected to a Canberra DSA-LX multichannel analyzer integrated into a microcomputer available from the LAN laboratory as shown in figure 1. Gamma ray spectra were collected and processed using the Genie 2000 software.



(a) Gamma spectrometry system



(b) Schematic representation, out of scale, of the experimental arrangement for measurements with the gamma spectrometer (Canberra 8).



(b) Representation of some standard radioactive sources used in the present work.

Figure 1: Experimental arrangement

The efficiency curve for the spectrometer was determined at a “reference” position, at approximately 100 mm source to detector distance, in order to make true-coincidence effects negligible, using standard sources of ^{133}Ba , ^{60}Co , ^{137}Cs , ^{152}Eu and ^{22}Na , as shown in table 1, with energies ranging from 121 keV to 1408 keV and applying the following equation:

$$\varepsilon_p(E) = \frac{N_p}{I_\gamma A_0 t_m DC} \quad (1)$$

where:

N_p is the area under the total energy absorption peak for the considered energy;

I_γ is the absolute gamma ray emission intensity of the line;

A_0 is the source activity;

t_m is the measurement time;

D is the decay factor - correction in the activity of the radioactive source for the acquisition date of the gamma spectrum. t_d is the decay time;

C is the decay factor during counting - correction for the decay during the measurement time t_m .

Table 1: Nuclear sources certificates.

Radionuclide	Source nr	Certificate nr	A_0 (kBq)	σ (%)	Calibration date
^{133}Ba	146S15	C/044/A15	15.12	1	5/10/15 12:00 PM
^{60}Co	144S15	C/042/A15	4.079	1.1	2/10/15 12:00 PM
^{137}Cs	147S15	C/045/A15	13.839	1.4	5/10/15 12:00 PM
^{152}Eu	52024	Al-010/1992	402	3.0	1/02/92 12:00 AM
^{22}Na	22S09	C/40/A09	71.5	3.5	19/06/09 12:00 AM

The corrections for the cascade summing effect, in this position, were assumed to be negligible. The target materials used as monitors were ^{198}Au , ^{95}Zr and ^{97}Zr the relevant nuclear data of these monitors can be seen in Table 2.

Table 2: Monitors and relevant nuclear data[10].

Index	Reaction	E_r (eV)	Q_0	$T_{1/2}$	E(keV)	$k_{0,Au}$
1	$^{96}\text{Zr}(n, \gamma)^{97m}\text{Zr}$	338	251.6	16.74 h	743.3	1.24×10^{-5}
2	$^{94}\text{Zr}(n, \gamma)^{95}\text{Zr}$	6260	5.31	64.02 d	724.2 + 756.7	2.00×10^{-4}
3	$^{197}\text{Au}(n, \gamma)^{198}\text{Au}$	5.65	15.7	2.695 d	411.8	1

E_r : average resonance energy

Q_0 : is the ratio of the resonance integral (I_0) to the thermal neutron cross section (σ_0), that is, $Q_0 = \frac{I_0}{\sigma_0}$.

$T_{1/2}$: nuclide half-life.

E: gamma ray energy.

k_0 : is the factor k_0 for the analyzed isotope, with reference to the comparator (Au).

The monitors were placed in polyethylene bags, sealed and their masses were obtained using a Shimadzu model AEL-40Sm analytical balance (Shimadzu Corp., Kyoto, Japan), accurate to 0,00001g. Then, the monitors were placed in polyethylene "rabbits". The masses and geometries of the monitors are listed in Table 3.

Table 3: Mass of monitors (Au and Zr).

Monitor	Features	mass(g)
Au	IRMM 530 RC wire $\Phi = 1.0$ mm Alloy Al-0.1 % Au	0.00812
Zr	Aldrich foil 0.25 mm 99.8 %	0.04553

The monitors were irradiated simultaneously in the pneumatic station (short irradiation system) of the IEA-R1 reactor for 1 minute under a thermal neutron flux of $(1.90 \pm 0.15)10^{13} \text{ n cm}^{-2} \text{ s}^{-1}$.

The induced activity measurements after the irradiation of the monitors were performed using the following procedure, was considered 5 hours decay time, then was measured the Zr foil to get the activity of ^{97}Zr . After 3 or 4 days it was measured again for the activity of ^{95}Zr , this procedures are necessary because the half-life of each one, 16.74h and 64.02d respectively.

This procedure was performed twice, for two sets of different samples in order to obtain a better comparison and reproducibility of the results.

The α parameter value was determined by the bare triple monitor method, we also used the Monte Carlo method to calculate the uncertainty of this parameter. The ratio (f) between the thermal and epithermal neutron fluxes was determined by the bi-monitors method using the isotopes ^{95}Zr and ^{97}Zr .

3. RESULTS AND DISCUSSION

The efficiency curve for the HPGe spectrometer was determined using the standard sources and the efficiency curve in function of energy of the detector is shown in Fig.1.

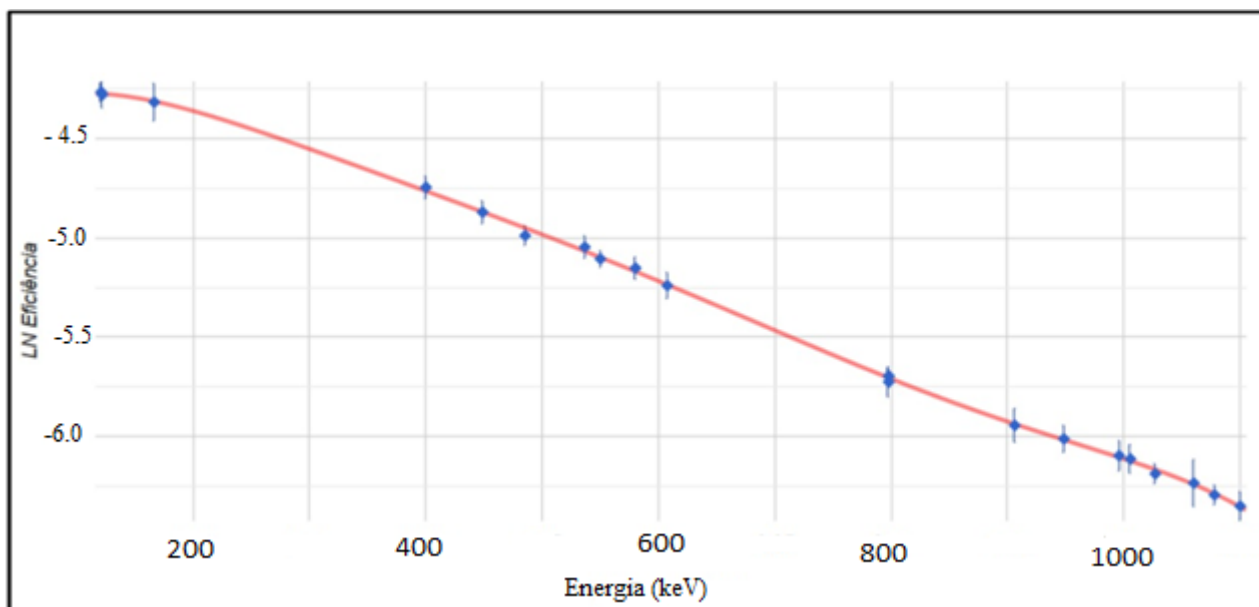


Figure 1: Efficiency curve obtained for the HPGe detector as a function of energy, using 100mm as distance source-detector.

The efficiency curve was adjusted by the least squares method, according to equation 2. The degree of the polynomial was 6 and the reduced Chi-Square resulted 0.989, indicating a satisfactory fit.

$$\ln(\varepsilon_p) = \sum_{i=0}^n a_i (\ln(E))^i \quad (2)$$

The parameter α was obtained through the "Bare Triple Monitor" [9] method using the Wolframalpha software and the Bisection Method [11]. The average value obtained was 0.0384 ± 0.0016 . The correction factor for self-shielding was considered and the Monte Carlo method was applied, in order to determine the uncertainty in α .

The parameter f was determined by the bi-monitors method. The value obtained was 35.67 ± 0.26 , indicating that this irradiation position is well thermalized.

In Table 4 there is a comparison of α and f values over time and for different detector, and it can be observed that the data are compatible with each other with a confidence level of 1 sigma, are reproducible, it is independent of the detector and the results depend only on the irradiation position.

Table 4: α and f values obtained for the pneumatic station of the IEA-R1 reactor.

Irradiation	Detector	α	f
October [13]2013	Canberra 1	0.0396 ± 0.0006	35.43 ± 0.23
November [14] 2015	Canberra 8	0.0384 ± 0.0014	35.67 ± 0.26
November [15] 2016	Canberra 3	0.0381 ± 0.0009	35.32 ± 0.19
Present work 2019	Canberra 8	0.0384 ± 0.0016	35.67 ± 0.26
Semmler [12] 2012	Canberra 1	0.043 ± 0.008	35 ± 2

4. CONCLUSION

The values of α and f were determined along a period of eight years and a reproducibility was observed in the results. The values obtained in this work are in agreement with those obtained by Semmler [12] according to the gauss normal distribution, but it is still necessary to verify the experimental parameters α and f obtained in this work by determining the element concentrations using some standard reference material.

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