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Influence of the Temporal Variability of Neutron Fluxes in the Carousel of TRIGA Mark IPR-R1 Research Reactor, CDTN, Brazil, in the k_0-Method of Neutron Activation Analysis

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ABSTRACT

The objective of this study was to verify quantitatively the temporal variation of neutron flux in the IC-40 of the carousel facility of the TRIGA reactor. It was determined experimentally that the flux variation was about 30% and 16% based on the operator's logbook during 8 hours of irradiation and equations were built to correct the flux variation in the k_0-method. The elemental concentrations of the certified reference material were corrected and showed that there was negligible influence of temporal variation in 8 hours' irradiation.

1 INTRODUCTION

Neutron Activation Analysis is one of the applications of the research nuclear reactor. This analytical technique is based on measurement of gamma rays emitted by radionuclides produced during irradiation of chemical elements in a material. The nuclear process is by neutron capture of the atom parent. The rate of gamma ray's emission is proportional to the concentration of the element [1]. The nuclear reaction with neutrons takes place in the nucleus, therefore there is no interference with the electrons in the orbitals. Thus, it is a purely elemental analysis, not determining the chemical species of the target element. This analytical technique determines chemical elements in several matrixes, in a large range of concentration (from traces to percentage) and high accuracy. The most efficient way to apply the technique is through the k_0-standardization method [1]. The method is based on four "pillars": data of the spectral parameters of irradiation channel of the reactor carefully determined, the gamma spectrometer absolutely calibrated, constants available in the literature and data of the sample under study.

The k_0-method is applied to determine chemical elements through short, medium and long half-life radionuclides in various types of matrices in steady-state operation mode of research reactors. It considers that the irradiation process is constant and continuous. It means that in long term irradiation, the neutron fluxes variation is not taken in account and it produces a systematic error in the results [2].

The k_0-standardization method has been applied at Laboratory for Neutron Activation Analysis (LNAA), Nuclear Technology Development Centre, located in Belo Horizonte, Minas Gerais state, Brazil, since 2003 [3]. The 100-kW nuclear research reactor TRIGA MARK I IPR-R1 has enabled the method to be carried out determining the elemental concentration of different matrices like soil, sediment, food, human tissues, etc. meeting requests of customers of CDTN and other institutions. The method has also been applied in development of researches at LNAA.

Over the years, the Laboratory for Neutron Activation Analysis has invested in improvement of the k_0-method. Therefore, the subject of this paper is a step forward to reach better analytical results. The time-dependent variation was measured in carousel facility of the TRIGA reactor during the long irradiation operation and it was proposed a factor to correct this.

2 TRIGA REACTOR OPERATION

The configuration of the core of TRIGA - IPR-R1, Figure 1, is to operate at 250 kW, however, the reactor still licensed to operate at nominal power of 100 kW. During the irradiation, the power control of the TRIGA IPR-R1 reactor is done by using ionization chambers. The information that the chambers send to the reactor control board is deformed due to the movement of the control bars during operation. The movement of the regulating control rod compensates the reactivity change caused by temperature and xenon build up, consequently occurs the redistribution of spatial neutron flux. The power of the TRIGA reactor is controlled and reestablished by operators [2].

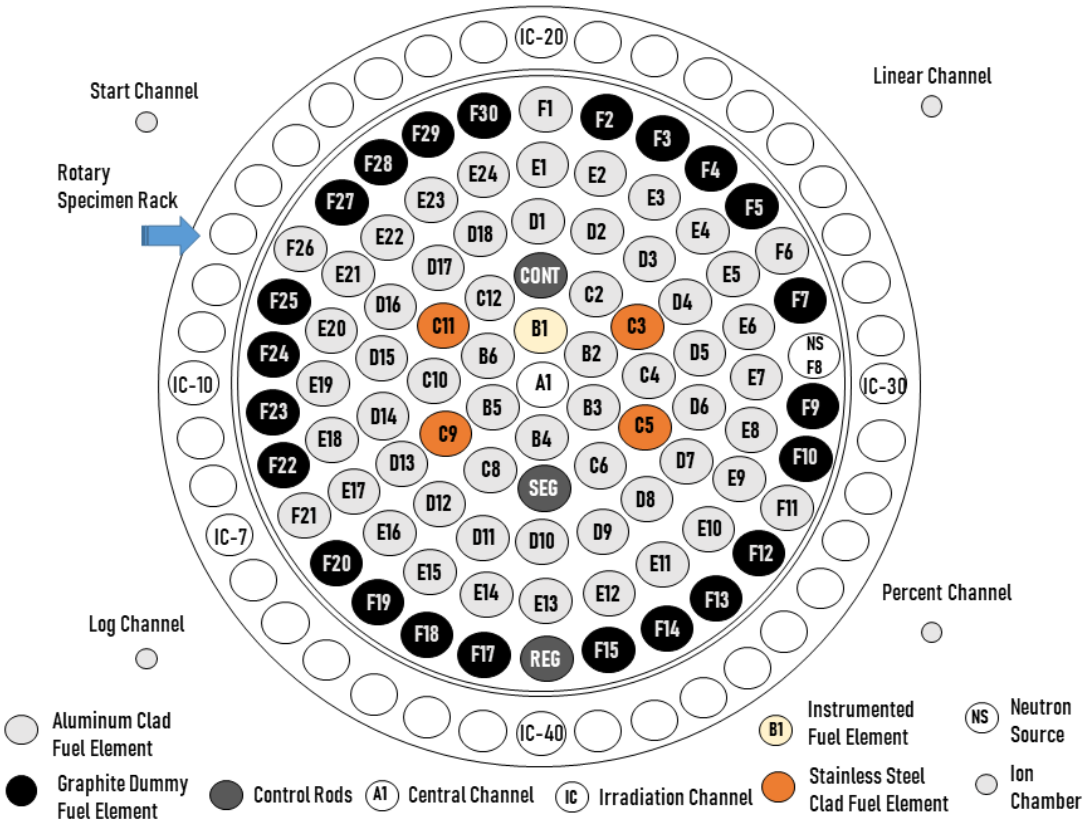

Figure 1: Core of the research nuclear reactor TRIGA Mark I IPR-R1 [4]

3 k_0-STARDARDIZATION METHOD

The basic equation of the k_0-method [3] for elemental concentration calculation for an analyte (a), ρ_a, is:

$$\rho_a = \frac{\left(\frac{N_p/t_m}{S\,D\,C\,W}\right)_a}{\left(\frac{N_p/t_m}{S\,D\,C\,w}\right)_{Au}} \times \frac{1}{k_{0,Au}(a)} \times \frac{G_{th,Au}\,f + G_{e,Au}\,Q_{0,Au}(\alpha)}{G_{th,a}\,f + G_{e,a}\,Q_{0,a}(\alpha)} \times \frac{\varepsilon_{p,Au}}{\varepsilon_{p,a}} \quad (1)$$

Where a is the analyte, Au refers to the co-irradiated gold monitor [^{197}Au(n,γ)^{198}Au, E_γ = 411.8 keV] and N_p is the net number of counts in the full-energy peak (corrected for pulse losses), W is the mass of the sample, w is the mass of the gold monitor, t_m is the measuring time, $S = 1\text{-}exp(-\lambda\, t_{irr})$, λ is the decay constant, t_{irr} is the irradiation time, $D = exp(-\lambda\, t_d)$, t_d is the decay time, $C = (1\text{-}exp(-\lambda\, t_m))/(\lambda\, t_m)$, f is the thermal to epithermal neutron fluence rate ratio, $Q_0 = I_0/\sigma_0$ (resonance integral to 2200 m s^{-1}/cross-section), α is the measure for the epithermal neutron fluence rate distribution, approximated by a $1/E^{1+\alpha}$ dependence (with a considered to be independent of neutron energy), and ε_p is the full-energy peak detection efficiency. G_{th} and G_e are factors for correction of thermal and epithermal neutron self-shielding, respectively.

In Eq. (1), the $k_{0,Au}$ is defined as:

$$k_{0,Au}(a) = \frac{M_{Au}\;\theta_a\;\sigma_{0,a}\;\gamma_a}{M_a\;\theta_{Au}\;\sigma_{0,Au}\;\gamma_{Au}} \quad (2)$$

where M is the atomic mass, θ is the isotopic abundance, σ_0 is the 2200 m s^{-1} (n,γ) cross-section, and γ is the absolute gamma-ray intensity (emission probability). The $k_{0,Au}$ is a constant experimentally measured and published in reference literature [5] and it is independent with neutron spectrum.

The detection of parameters involved in activity determination by gamma-ray spectrometry, including both the radionuclide of interest (index a) and comparator (index Au) are related to the first and forth factors in Eq. (1). The second factor is k_0-factor and the third factors are related to parameters involved with neutron field characteristics of irradiation channel (f and α) and $Q_0(\alpha)$.

According to Jaćimović and co-authors, 2012, [2] when the neutron flux changes and the spectral parameters f and α continue constant, the expression for S, that is $S = 1\text{-}exp(-\lambda\, t_{irr})$, has to be placed by:

$$S' = \int_0^{t_{irr}} F(t)\;\lambda\; e^{\lambda(t-\,t_{irr})}\,dt \quad (3)$$

In this expression, $F(t)$ is a function of the time-dependent neutron flux, normalised in order it equals 1 in the irradiations zero-time. It is important to mention that the correction of the saturation factor (S0) depends on the half-life of the nuclide and the irradiation time.

One approach is linear time dependence of the neutron flux can be expressed as:

$$F(t) = 1 + k\;t \quad (4)$$

In this Eq. (4), k is the slope of neutron flux during irradiation.

Another relevant factor is $F_{c,Au}$-factor, Eq. (5), called comparator factor, calculated based on neutron monitors irradiated together with the sample. The usual is to irradiate the sample intercalated by the monitors. This factor shows the trend of the axial neutron flux gradient, while the radial gradient is negligible due to similar diameter of sample and standard (Al-0.1%Au alloy).

$$F_{c,Au} = \frac{\emptyset_{th}}{f} \times \frac{1}{3.47E+6} \quad (5)$$

The $F_{c,Au}$ - factor, as defined in Eq. (5), is proportional to the thermal neutron flux density and directly indicates a gradient in epithermal flux density; $f\left(\frac{\emptyset_{th}}{\emptyset_e}\right)$ is the ration between thermal and epithermal fluxes. This factor can be calculated by KayWin software based on the irradiated Al-0.1%Au monitors.

4 METHODOLOGY

In this study, the irradiation channel IC-40 in the carousel facility of the TRIGA IPR-R1 reactor was chosen to measure the temporal variation of the neutron flux during long irradiation, usually 8 hours. The reason is the compensating rod is not only lined up radially with this IC and also close. Four sets of monitors, Au, were irradiated to verify the temporal variation.

4.1 Preparation of sets of monitors

Monitors of Al-0.1%Au foil, IRMM-530R alloy (6 mm diameter-disc and 0.1 mm high, (0.1003 ± 0.0012) % purity of Au with k=2) were used. Before irradiation they were cleaned with alcohol and deionized water, dried and weighted in analytical balance. Each set monitor was inserted in a polyethylene vial and then, in the rabbit.

4.2 Irradiation of sets of monitors

The irradiations were carried out in IC-40 in the carousel facility of the 100 kW TRIGA MARK I IPR-R1 reactor. After the reactor was critical at 9:01 h, each rabbit was inserted and taken manually out without reactor stopping. Each rabbit was irradiated one by one and the irradiation time was controlled and written in the logbook as follows: monitor 1, from 9:07:23 to 9:27:26; monitor 2, from 11:07:00 to 11:27:06; monitor 3, from 13:07:00 to 13:27:00 and monitor 4, from 16:37:00 to 16:57:00.

4.3 Gamma spectrometry of sets of monitors

The induced activities were measured after suitable decay time on an absolute calibrated HPGe detector, GC 5019, CANBERRA, with 50% relative efficiency. The spectra were acquired with Genie 2000 software, CANBERRA. The HyperLab program (HyperLabs software, Budapest, Hungary, 2002) was used for peak area evaluation and the software package Kayzero for Windows® (User's Manual, for reactor neutron activation analysis (NAA) using the k_0-standardization method, ver. 3.37), also called KayWin, was applied. This is a specific program to calculate the elemental concentration and it also calculates the specific activity (A_{sp}).

4.4 Verification of temporal variation

To check the neutron flux changes for 8 hours irradiation, one procedure was carried out to determine the Specific Activity of the Au monitor. Table 1 shows the relevant nuclear data used for calculations.

Table 1: Measurements of the monitors at CDTN

Monitor	Nuclide	Reaction	$T_{1/2}$ (unc, %)	E_γ, keV	Measurement (Distance monitor-detector)
Au	^{198}Au	^{197}Au(n,γ)^{198}Au	2.695 d (0.008)	411.8	10 cm

5 RESULTS AND DISCUSSION

The values for A_{sp} calculated for each Au monitor, based on gamma measurement, are displayed in Table 2. It shows a decreasing trend based on A_{sp} – monitor 1, 2 and 3 – however the values have increased for the last monitor 4, irradiated at the end of operation. This result suggests that the operator has interfered and repositioned the control bars to keep the power at 100 kW. Figure 2 shows the experimental trend of neutron flux in function of irradiation time. The slope of the curve points out that the reactor's power decreases -3.8169% per hour.

Table 2: Values for A_{sp}

Monitor	Start of Irradiation	End of Irradiation	^{198}Au Measurement	A_{sp} (cpm)	Power (%)	Difference (%)
-	09:01*	-	-	-	-	-
1	09:07:23	9:27:26	AU1	2.95E+11	100.00	0
2	11:07:00	11:27:06	AU2	2.88E+11	97.62	-2.38
3	13:07:00	13:27:00	AU3	2.72E+11	92.37	-7.63
4	16:37:00	16:57:00	AU4	3.01E+11	100.00	0
-	17:01**	-				

Note: *, reactor critical; **, Scram; A_{sp}, Specific Activity; Difference is related to 100%

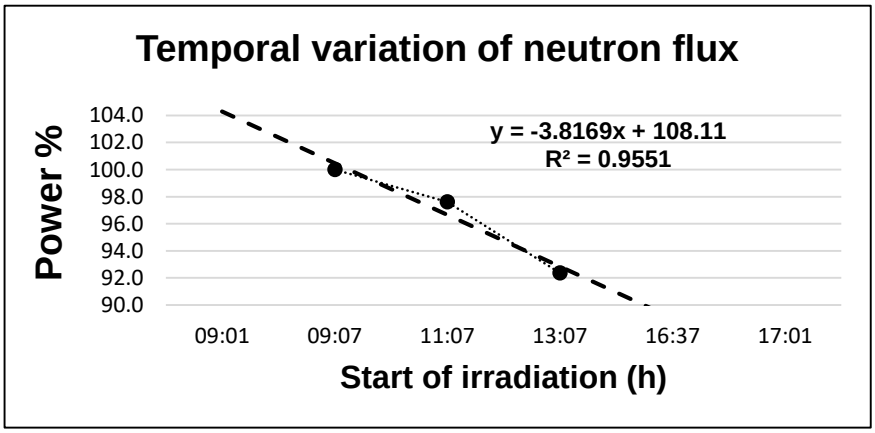

Figure 2: Experimental values showing the decreasing trend of neutron flux in function of irradiation time

In order to complement the information about the temporal variation, the operators made available the power variation notes during the irradiation of the certified reference material BCR-320R, Channel Sediment (European Commission's Joint Research Centre, Institute for Reference Materials and Measurements, Belgium) for 8 hours. Table 3 displays the values written in the logbook and Figure 3 shows the curve of the trend. This curve points out that the reactor's power decreases -1.9167% per hour.

Table 3: Variation of reactor's power in 8 hours irradiation time

TRIGA IPR-R1		
Irradiation Time	**Time (h)**	**Power (%)**
10:08:59		0
10:09:00	0	100
11:09:00	1	99
12:09:00	2	97
13:09:00	3	95
14:09:00	4	93
15:09:00	5	91
16:09:00	6	89
17:09:00	7	87
17:59:00	8	100

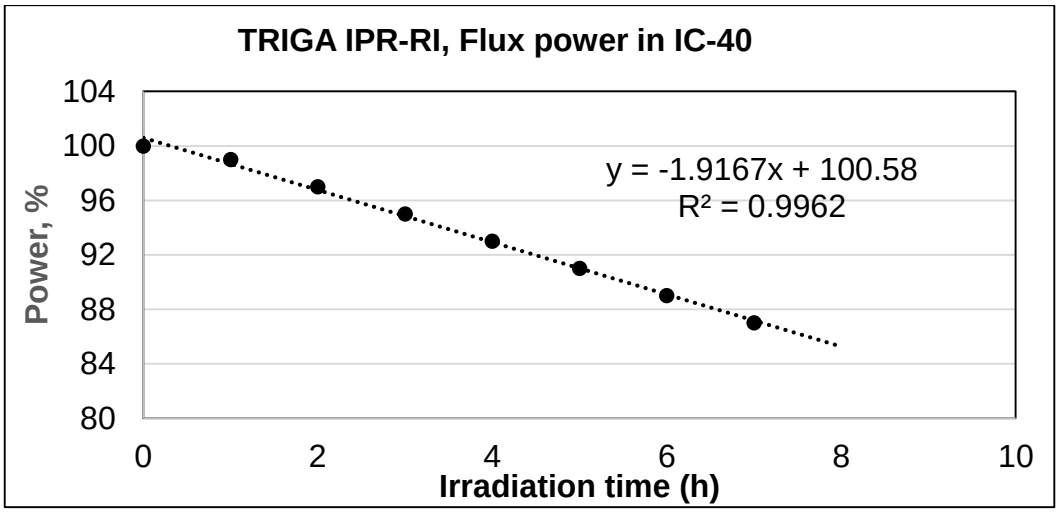

Figure 3: Notes from operator's logbook showing the decreasing trend of neutron flux in function of irradiation time

In order to evaluate the contribution of the variation temporal in the k_0-method calculations during long irradiation time, two files were prepared to correct the values. These files were based on slope of the curves that describe the decreased trend, one called ES, Experimental Slope and other S, Slope, based on records in the logbook.

The software KayWin apply a file *.FLX to make the corrections according to the irradiation time. Several elemental concentrations were determined, however, this paper will

consider only the elements that have certified values. The certified reference material BCR-320R had the elemental concentrations calculated without variability of time corrections, with corrections made by ES file and corrections made by S file. The corrections were made by correcting the value of Fc using ES and S files. Table 4 shows the nuclear data used to calculate the elemental concentrations in the sample.

To evaluate the results, the E_n score [6] were applied to certified values as well as Relative Bias (%). The E_n considers the expanded uncertainty of the experimental analysis and the certified values with a coverage factor $k = 2$ (95% confidence interval). The evaluation criteria points out that the performance is considered satisfactory when is $|E_n| \leq 1$, and unsatisfactory when and $|E_n| > 1$. When $|E_n| \leq 1$, it means that the method produced results with 95% of possibility to be inside a range of values that correspond to the true values. Table 5 shows the all values are $|E_n| \leq 1$.

Table 4: Nuclear data used in the KayWin software to calculate the mass fractions of CRM BCR-320R

El.	Nuclide	$T_{1/2}$	E_γ, keV
As	^{76}As	26.24 h	559.1
Co	^{60}Co	5.275 y	1173.2; 1332.5
Cr	^{51}Cr	27.7 d	320.1
Fe	^{59}Fe	44.5 d	1099.3; 1291.6
Sc	^{46}Sc	83.83 d	889.3; 1120.5
Th	^{233}Pa	26.97 d	311.9
U	^{239}Np	2.357 d	228.2; 277.6
Zn	^{65}Zn	244.3 d	1115.5

Table 5: Evaluation of experimental results by Relative bias (%) and E_n-*score* for the certified values of BCR-320R (mg/kg)

El.	Certified Values, k = 2	Without Corrections Fc_{ave} = 8770, (k =1)		Correction ES, -3.8169% /h Fc_{ave} = 8788, (k = 1)			Corrections S, -1.9167% /h Fc_{ave} = 8779, (k = 1)		
		Experimental Values	E_n-*score*	Experimental Values	Rel. Bias, %	E_n-*score*	Experimental Values	Rel. Bias, %	E_n-*score*
As	21.7 ± 2.0	22.5 ± 0.8	0.33	22.6 ± 0.8	0.22	0.35	22.6 ± 0.8	0.04	0.33
Co	9.7 ± 0.6	9.68 ± 0.34	-0.03	9.66 ± 0.34	-0.21	-0.05	9.67 ± 0.34	-0.10	-0.04
Cr	59 ± 4	60.1 ± 2.1	0.19	60.0 ± 2.1	-0.18	0.18	60.1 ± 2.1	-0.08	0.19
Fe	25700 ± 1300	24940 ± 875	-0.35	24890 ± 873	-0.20	-0.37	24910 ± 874	-0.12	-0.36
Sc	5.2 ± 0.4	5.19 ± 0.18	-0.02	5.18 ± 0.18	-0.19	-0.04	5.19 ± 0.18	-0.10	-0.03
Th	5.3 ± 0.4	5.16 ± 0.19	-0.26	5.15 ± 0.19	-0.19	-0.28	5.16 ± 0.20	0.04	-0.25
U	1.56 ± 0.20	1.56 ± 0.06	-0.01	1.56 ± 0.06	0.00	-0.01	1.56 ± 0.06	0.00	-0.01
Zn	319 ± 20	318 ± 11	-0.02	318 ± 11	-0.22	-0.05	318 ± 11	-0.09	-0.03

Fc_{ave}, Average of Correction Factor

6 CONCLUSIONS

Experimental evaluation and information based on logbook of the reactor's operator showed that the neutron flux varies in function of irradiation time, -3.8169% per hour and -1.9167% per hour, respectively. However, the results for certified reference material BCR-320R irradiated for 8 hours pointed out that the temporal variability is negligible to the studied certified values. It is suggested that the variability is inside the uncertainty of the results, even for medium and long half-live radionuclides. In conclusion, for samples irradiated for 8 hours, the elemental concentrations results will be similar with or without corrections related to variation of neutron flux up to 30%.

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