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Characterization of irradiation channels in the carousel of TRIGA Mark I IPR-R1 research reactor, Brazil, aiming at the application of k_0-standardization method of neutron activation analysis

Maria Ângela de Barros Correia Menezes[1] · Daniel de Almeida Magalhães Campolina[2] · Radojko Jaćimović[3]

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Abstract

New values of neutron fluxes and spectral parameters f and α were determined experimentally in all irradiation devices of the TRIGA Mark I IPR-R1 nuclear research reactor at Nuclear Technology Development Centre (CDTN), Belo Horizonte, Brazil. Sets of monitors Au, Fe, Zn and Zr were irradiated bare and Cd-covered, according to "Cd-ratio for multi-monitor" method. Values were validated by analysing the certified reference material BCR-320R irradiated in chosen channels. The calculations were made based on irradiation channel values and the average values of the Carousel. The results of E_n-score point out that the k_0-method is producing reliable results. From now on, the values of mass fractions in several matrices, the production and studies with radioisotopes will be more accurate and the activities calculated more precisely.

Keywords Spectral parameters · Neutron fluxes · Validation · Reference sample · BCR-320R · Kayzero for Windows

Introduction

The Nuclear Technology Development Centre, Brazilian Commission for Nuclear Energy, CDTN/CNEN, celebrates 70 years of the first criticality of its TRIGA Mark I IPR-R1 research reactor in 2022 [1]. This nuclear reactor is a multipurpose tool built by General Atomic for training, research, isotopes [2]. It is a unique and pioneering facility in Brazil and it was acquired by the government of the State of Minas Gerais.

Along the time, the reactor has been used for several objectives like elemental concentration determination of samples of various matrices by neutron activation analysis (NAA), research of new substances with potential uses in health and radiopharmacy, production of radioisotopes for industrial applications and tracers for environmental studies and neutronic and thermohydraulic studies. Related to training, the Research Reactor Operator Training Course (CTORP) was a pioneer in the training of operators at Angra's nuclear power plants.

The TRIGA IPR-R1 reactor has three irradiation devices in use: the Central Thimble, the Rotary Rack and the Fast Pneumatic Transfer Terminal 2 [3]. The Central Thimble allows the irradiation of samples in the position where the neutron flux is maximum, the largest contribution being from fast neutrons, followed by epithermal and thermal.

The Rotary Rack or Carousel consists of a ring that surrounds the core with 40 channels or positions for sample irradiation. The access of the samples, inserted in the sample holder, to the irradiation positions, is through a cable that goes down an access tube. It is the most requested location for irradiation, as the component of thermal neutrons is the one that most contributes to the total flux, followed by epithermal and fast ones. For activations in general, a more intense flux of thermal neutrons is interesting. Furthermore, more samples can be irradiated simultaneously.

✉ Maria Ângela de Barros Correia Menezes
menezes@cdtn.br

1 Nuclear Technology Development Centre / Brazilian Commission for Nuclear Energy, CDTN/CNEN; Division for Analysis and Environment, Laboratory for Neutron Activation Analysis., Av. Pres. Antônio Carlos nº 6627, Campus UFMG, Belo Horizonte, Minas Gerais 31270-901, Brazil

2 Nuclear Technology Development Centre / Brazilian Commission for Nuclear Energy, CDTN/CNEN, Division of Unit of Reactor TRIGA., Av. Pres. Antônio Carlos nº 6627, Campus UFMG, Belo Horizonte, Minas Gerais 31270-901, Brazil

3 Jožef Stefan Institute, Department of Environmental Sciences, Jamova cesta 39, SI-1000, Ljubljana, Slovenia

The Fast Pneumatic Transfer Terminal 2 (FPTT2) consists of a system that allows the fast introduction and withdrawal of samples. This terminal is mainly used for the analysis of uranium by the delayed fission neutron method [4]. As it is outside the core (Fig. 1), it is the most thermalized irradiation position of the TRIGA reactor.

Along the years, the core configuration of the TRIGA reactor was modified six times: in 1964 (configuration n. 2), 1967 (configuration n. 3), 1973 (configuration n. 4), 1996 (configuration n. 5), and 2001 (configuration n. 6). Diversified experimental and semi theoretical methodologies as Monte Carlo Code [5] have been determining the neutron fluxes in different irradiation channels and devices, applying different procedures and materials [6–17].

Table 1 shows that the measurements of neutron fluxes were carried out in different places and by different procedures, also using different materials related to the reactor core configuration changings. Over time, studies were carried out to characterize the neutron fluxes in their irradiation devices. Table 2 shows the averaged values of thermal and epithermal neutron fluxes determined by different authors for the core configurations number 5 and 6 of the IPR-R1 reactor at 100 kW, related to Carousel and Central Thimble.

Relative method of neutron activation analysis started to be applied just after the first criticality and it was the main method to determine elemental concentrations up to 1995 when the k_0-method was established.

Until 2001, the carousel of the reactor was used to rotate. Due to this, average values for thermal and epithermal fluxes were used whenever necessary. However, while the core was changed for a new configuration to enable the operation from 100 kW to 250 kW, configuration n. 6, a damage was observed in the mechanism to rotate. Since then, the carousel no longer rotates during irradiations preserving the damaged mechanism of the carousel. It means that from this time on, it was necessary to determine the neutron fluxes and f (thermal-to-epithermal neutron flux ratio) and α (epithermal neutron flux shape factor) in all irradiation channels since it was no longer possible to use the average values. In the application of k_0-method, it was necessary to guarantee accuracy in the application of the method. The k_0-method was re-established and improved in 2003 when five specific irradiation channels in the carousel were chosen to have the spectral parameters determined again [13].

Nevertheless, the other irradiation channels (Carousel and Central Thimble) went on to be used by other projects for production of radioisotopes and radiopharmaceuticals and the calculations of activities were performed using

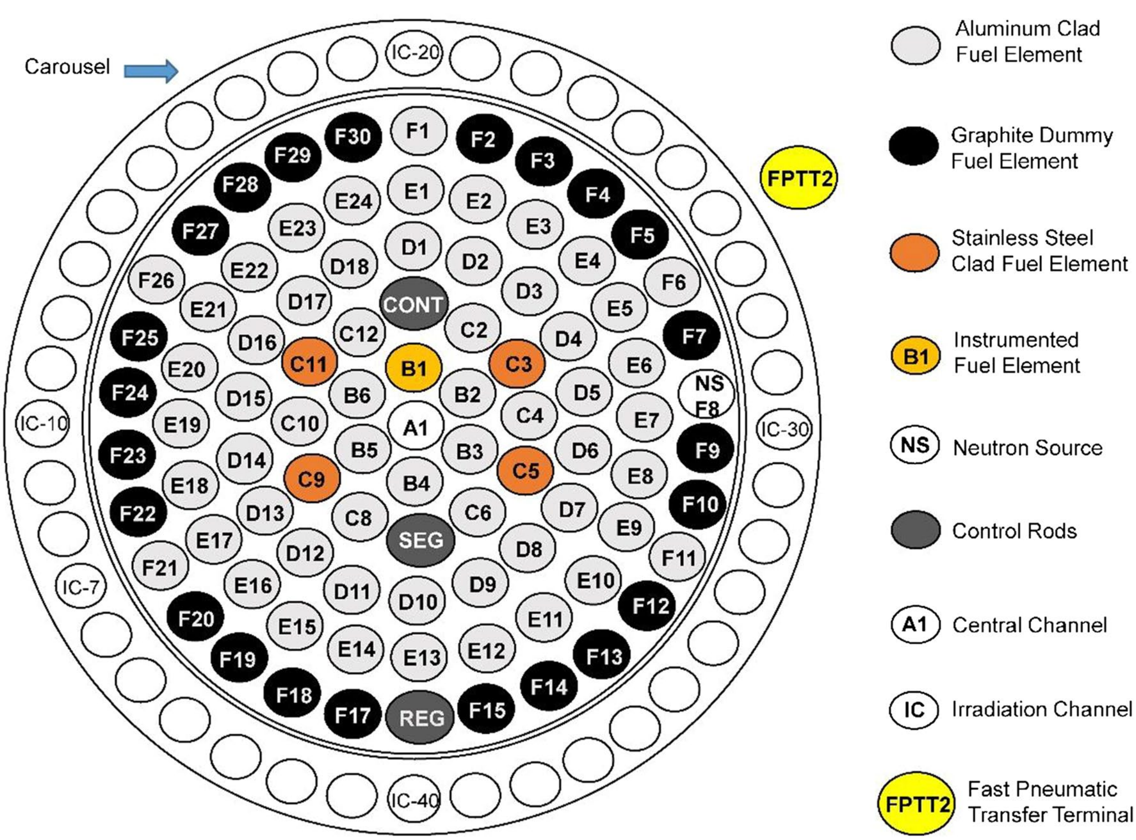

Fig. 1 Core configuration n. 6 of TRIGA Mark I IPR-R1 research reactor

Table 1 Measurements of neutron fluxes along the years

References	Core Configuration number	Description	Irradiation device	Material used in the experiments
Santoro [7]	3	Experimental determination: Characterization of epithermal and fast neutrons	Central Tube	Reference materials: In, Au, Mn, Na, Ni, Al, Th
Guimarães [8, 9]	4	Experimental determination: axial profile of fast and thermal neutrons	Some irradiation channels	Sensors on stainless steel electrodes
Dalle [6]	5	Theoretical determination: thermal flux by computational codes: MCNP4B, ORIGEN2.1 and MONTEBURNS	Central Tube and Carousel	–
Sousa [12]	5	Experimental determination: average thermal flux	Central Tube and Carousel	Au foil, Cd-box
Franco and Sabino 10, Franco, [11]	5	Experimental determination: thermal and epithermal fluxes and spectral parameters f and α	Central Tube and Carousel (average values)	ZrO_2, Zr metal, LuO_2 (Johnson Matthey Chemicals Ltda.), solution of Au (NBS)
Menezes and Jaćimović [13]	6	Experimental determination: thermal and epithermal fluxes and spectral parameters f and α, "Cd-ratio for multi-monitor" method [18].	Carousel (five specific irradiation channels)	Al– 0.1%Au alloy, IRMM-530R; Zr 99.8% Goodfellow; Fe ≥ 99.996%, IRMM-524 A; Zn 99.99+%, Goodfellow; Cd-box (Goodfellow)
Zangirolami et al. [14]		Experimental determination: thermal and epithermal neutron fluxes	Carousel, Central Tube and Pneumatic Fast Transfer 2	Al– 0.1%Au alloy, IRMM-530R; Cd-box
Guerra [15]		Theoretical determination: thermal fluxes in each irradiation channel and average value by computational code Monte Carlo	Carousel (11 irradiation channels)	–
Salomé et al. [16]		Theoretical determination: thermal fluxes in each irradiation channel and average value by computational code Monte Carlo	Carousel (11 irradiation channels)	–
Jaćimović and Menezes [17]		Experimental determination: thermal and epithermal neutron fluxes "Cd-ratio for multi-monitor" method [18]	Carousel, IC-7	Al– 0.1%Au alloy, IRMM-530R; Zr 99.8% Goodfellow; Fe ≥ 99.996%, IRMM-524 A; Zn 99.99+%, Goodfellow; Cd-box (Goodfellow)

Table 2 Neutron fluxes determined for core configurations numbers 5 and 6 of TRIGA IPR-R1 reactor, Carousel and Central Thimble

References	Core configuration number	Carousel (average value)		Central Thimble (average value)	
		Thermal Flux 10^{11} cm^{-2} s^{-1}	Epithermal Flux 10^{10} cm^{-2} s^{-1}	Thermal Flux 10^{12} cm^{-2} s^{-1}	Epithermal Flux 10^{11} cm^{-2} s^{-1}
Souza [12]*	5	6.6 ± 0.4	–	4.1	–
Franco and Sabino [10], Franco [11]		5.9 ± 0.1	2.5 ± 0.2	2.9 ± 1.0	1.6 ± 0.4
Dalle [6]**	6	7.2	–	5.2	–
Menezes and Jaćimović [13]*		6.3 ± 0.6	2.9 ± 0.3	–	–
Zangirolami et al. [14]*		8.1 ± 0.3	3.4 ± 0.2	2.8 ± 0.1	2.6 ± 0.1
Guerra [15]**		6.6 ± 0.5	–	–	–
Salomé et al. [16]**		6.5 ± 0.5	–	–	–

*, experimental determination; **, determination by simulation; -, not determined

values of average neutron fluxes. Values that needed to be re-determined.

This paper is about the determination of thermal, epithermal and fast neutrons and also spectral parameters in the irradiation channels of Carousel used to analyse the samples by k_0-standardized method of neutron activation analysis. The Carousel is related to core configuration n. 6, Fig. 1. The neutron fluxes and spectral parameters were also determined in F12 in the core (where the Fast Pneumatic Transfer Terminal 1 will be installed), in the Fast Pneumatic Transfer Terminal 2 (where irradiations to determine uranium by Delay Fission Neutrons are carried out) and in the Central Thimble as additional information.

Methodology

Determination of neutron fluxes and spectral parameters

The procedure to determine all data necessary for the k_0-method concerning the irradiation channel, consisted of two steps: (1) to determine the spectral parameters *f* (ratio thermal to epithermal neutron fluxes) and α (measure of the deviation of epithermal neutrons from the ideal 1/E distribution) by "Cd-ratio for multi-monitor" method [18] and (2) to calculate the values of the thermal, epithermal and fast neutron fluxes using specific equations, *f* and α already determined and spreadsheet help. The nuclear data used were according to the recommended k_0 database [19].

Step (1) Determination of spectral parameters according to "Cd-ratio for multi-monitor" method [18].

This method is the most accurate procedure to determine these parameters. In this step, sets of monitors were used. They were discs of 6 mm in diameter: Au (Al– 0.1%Au foil, IRMM-530R alloy, height of 0.1 mm and (0.1003 ± 0.0012) % purity); Fe (IRMM foil, IRMM-524 A, height of 0.1 mm, ≥ 99.996% purity); Zn (Goodfellow foil, height 0.050 mm, 99.99% purity) and Zr (Goodfellow foil, height of 0.125 mm, 99.8% purity) that were irradiated bare and Cd-covered (Goodfellow Cd rod, height of 20 mm, diameter of 10 mm, thickness of 1 mm, 99.99% purity) in all irradiation channels.

The preparation of monitors, procedure to irradiate and measurements of the induced activities were carried out according to procedure described by Jaćimović and Menezes, 2022 [17].

The software package called Kayzero for Windows® (KayWin) [20] was used to calculate parameter α by Cd-ratio for multi-monitor method and α was obtained as the slope (-α) of the straight line when plotting:

$$log\frac{\left(\bar{E}_{r,i}\right)^{-\alpha}}{\left(F_{Cd,i}\cdot R_{Cd,i}-1\right)\cdot Q_{0,i}(\alpha)\cdot\frac{G_{e,i}}{G_{th,i}}}\quad \text{versus}\quad log\bar{E}_{r,i} \tag{1}$$

i correspond to the isotope 1, 2, 3, …N.

Using the same software KayWin, *f* is also calculated:

$$f = Q_{0,r}(\alpha)\left(F_{Cd,r}R_{Cd,r}-1\right)\frac{G_{e,r}}{G_{th,r}} \tag{2}$$

Each monitor set was irradiated Cd-covered and bare conforming to Cd-ratio method. Therefore, the Cd-ratio (R_{Cd}) is determined (the ratio of specific activities of the bare and Cd-covered monitors), small corrections for Cd-transmission for epithermal neutrons (F_{Cd}) were applied as well as corrections for self-shielding of thermal and epithermal neutrons (G_{th}, G_e). Due to the unmanageable correction for G_e with the KayWin program, which takes into account the value of G_e=1, we calculated it using the MATSSF program [21] and used it in the calculation of the spectral parameters. Index *r* is the monitor with accurate known nuclear constants, mainly Q_0 and $\bar{E}_r$. Defined in this way, parameter *f* is the ratio of thermal and epithermal fluxes according to the Høgdahl convention (criterion $E_{Cd} \approx 0.55$ eV) [18]. The use of chosen monitor set neutron energy range from 5.65 eV (^{197}Au) up to 6260 eV (^{94}Zr) can be covered.

Step (2) Determination of thermal, epithermal and fast neutron fluxes.

To determine experimentally thermal and epithermal neutron fluxes, an Excel sheet based on f and α values determined previously using Cd-ratio for multi-monitor method was used. The Eq. 3 was applied to calculate the values for thermal neutron fluxes (ϕ_{th}):

$$\varnothing_{th} = \frac{\left(A_{sp}/M\right)}{\left(\mathbf{N_A}\theta\gamma\left(G_{th}\sigma_0 + G_e I_0(\alpha)/f\right)\varepsilon_p\right)} \tag{3}$$

where:

A_{sp} is specific count rate, M is molar mass, N_A is Avogadro number, θ is isotopic abundance, γ is absolute gamma intensity, G_{th} and G_e are correction factors for thermal and epithermal neutron self-shielding, respectively, and ε_p is full-energy peak detection efficiency. Epithermal neutron fluxes (ϕ_e) were calculated as a ratio between ϕ_{th} and f ($\phi_e = \phi_{th}/f$).

Equation 4 was used to calculate the values for fast neutron flux (ϕ_f) based on the same experiment in which f and α were determined using Cd-ratio for multi-monitor method.

$$\varnothing_f = \frac{\left(A_{sp}/M\right)}{\left(\mathbf{N_A}\theta\gamma G_f\sigma_f\varepsilon_p\right)} \tag{4}$$

where G_f is correction factor for fast neutron self-shielding and σ_f is cross section for fast neutrons. Usually calculated data for fast flux on this way are used due to low noise in monitors activation under Cd-covered. Based on chosen monitor set, fast flux can be calculated using $^{54}Fe(n,p)^{54}Mn$ reaction, which is a threshold reaction produced by median neutron energy ($E_{50\%}$) of 4.294 MeV [22]. Additionally, reaction $^{90}Zn(n,2n)^{89}Zr$ can be used, but due to high threshold energy ($E_{50\%}$=14.32 MeV [22]) may be inaccurate in case of well-thermalized neutron spectra as well as due to a spectrum interference for gamma line at 909.1 keV of ^{89}Zr by the 911.1 keV of ^{228}Ac from the background.

Validation

In our previous work, the validation of k_0-NAA has been demonstrated using different CRMs [23, 24]. In this work, the certified reference material BCR-320R [25] was analysed to validate the values determined in this study and to evaluate the analytical performance of the method. Aliquots of the certified reference material, about 200 mg, were weighed into polyethylene vials suitable for neutron irradiation. The samples were irradiated intercalated by neutron flux monitors (discs of Al− 0.1%Au alloy IRMM-530R, European Commission's Joint Research Centre, Institute for Reference Materials and Measurements (IRMM), Belgium, 6 mm diameter and 0.1 mm thick). The irradiation time was for 8 h in specific chosen irradiation channels in the carousel of the 100 kW TRIGA Mark I IPR-R1 reactor. The gamma spectrometry was carried out on an HPGe detector (50% relative efficiency, microcomputer, associated electronic CANBERRA) three times to measure radionuclides with medium and long half-lives (after 3–4, 7 and 21 days' decay time). For acquisition of spectra, the Genie 2000 software CANBERRA was applied. The spectra were deconvoluted by the HyperLab program [26] and the elemental mass fractions were calculated by a software package KayWin [20].

Statistical test

The E_n-score [27] test was applied to verify the analytical response of the k_0-method, which used the spectral parameters and neutron fluxes determined. The test verifies if the experimental results obtained for the certified reference material were consistent to certified values. The E_n-score considers the expanded uncertainty of the experimental analysis and the certified values with a coverage factor $k=2$ (95% confidence interval).

The following equations were used in the E_n-score calculations:

$$E_n = \frac{X_{Lab} - X_{Assigned}}{\sqrt{U_{Lab}^2 + U_{Assigned}^2}} \tag{5}$$

where U_{Lab} and $U_{Assigned}$ are the expanded uncertainties ($k=2$) of the experimental result (X_{Lab}) and the assigned result ($X_{Assigned}$), respectively, and

$$U_{Lab} = 2 \times u_{Lab\_Comb} \tag{6}$$

$$u_{Lab\_Comb} = \sqrt{u_{AREA}^2 + u_{method}^2} \tag{7}$$

where u_{AREA} is uncertainty of number of counts in the full-energy peak (AREA) and u_{method} is combined standard uncertainty ($k=1$) of k_0-NAA established at the CDTN/CNEN as 3.5%. This value is recommended according to De Corte, 1987 [18].

The evaluation criteria used is $|E_n| \leq 1.0$, showing that the performance was considered satisfactory. It means that the method produced results with 95% of possibility to be inside a range of values that correspond to the certified values. The performance is unsatisfactory when $|E_n| > 1.0$.

Results and discussions

The spectral parameters and neutron fluxes were determined for the irradiation channels (IC) of the Carousel from 2016 to 2019. For irradiation in channel IC-7, the values were

Table 3 Neutron fluxes and spectral parameters determined in all irradiation devices of TRIGA IPR-R1 reactor in 2016, 2018 and 2019 by using ^{198}Au, ^{59}Fe, ^{69m}Zn, ^{65}Zn, ^{95}Zr and ^{54}Mn

Channel	Neutron Fluxes			Spectral Parameters	
	Thermal Flux* 10^{11} cm^{-2} s^{-1}	Epithermal Flux* 10^{10} cm^{-2} s^{-1}	Fast Flux** 10^{10} cm^{-2} s^{-1}	f	α
IC– 1	7.53 ± 0.15	3.20 ± 0.06	7.92 ± 0.33	23.49 ± 0.89	0.00264 ± 0.1370
IC– 2	7.81 ± 0.28	3.49 ± 0.13	8.17 ± 0.34	22.39 ± 0.89	0.0108 ± 0.8899
IC-3	7.75 ± 0.24	3.32 ± 0.10	8.47 ± 0.37	23.49 ± 0.89	0.0026 ± 0.1370
IC-5	7.82 ± 0.0.16	3.56 ± 0.07	8.66 ± 0.36	22.05 ± 0.12	0.0036 ± 0.1345
IC-6	7.71 ± 0.28	3.34 ± 0.12	8.82 ± 0.37	23.08 ± 0.87	− 0.0074 ± 0.1336
IC-7^{+}	7.94 ± 0.27	3.71 ± 0.13	9.22 ± 0.39	21.42 ± 0.86	0.0039 ± 0.1328
IC-7R^{++}	7.78 ± 0.16	3.59 ± 0.07	9.06 ± 0.38	21.68 ± 0.86	−0.0001 ± 0.1328
IC-721#	7.97 ± 0.26	3.43 ± 0.11	8.56 ± 0.37	23.25 ± 0.09	− 0.0045 ± 0.1347
IC-8	7.60 ± 0.21	3.40 ± 0.09	8.79 ± 0.37	22.39 ± 0.86	− 0.0048 ± 0.1329
IC-9	7.77 ± 0.18	3.40 ± 0.08	8.47 ± 0.33	22.86 ± 0.87	− 0.0064 ± 0.1334
IC– 10	7.73 ± 0.20	3.39 ± 0.09	8.68 ± 0.37	21.78 ± 0.87	0.0021 ± 0.1336
IC– 11	8.00 ± 0.23	3.18 ± 0.09	9.43 ± 0.38	25.12 ± 1.10	− 0.0213 ± 0.1871
IC– 12	8.01 ± 0.16	3.62 ± 0.07	9.16 ± 0.38	22.15 ± 0.07	− 0.0021 ± 0.1332
IC– 13	7.93 ± 0.20	3.66 ± 0.09	8.96 ± 0.37	21.69 ± 0.08	− 0.0007 ± 0.1327
IC– 14	7.83 ± 0.25	4.06 ± 0.13	9.23 ± 0.39	19.29 ± 0.86	0.0201 ± 0.1331
IC– 15	8.07 ± 0.17	3.55 ± 0.07	9.12 ± 0.38	22.75 ± 0.87	− 0.0024 ± 0.1343
IC– 16	7.76 ± 0.22	3.40 ± 0.10	8.42 ± 0.36	22.79 ± 0.88	0.0011 ± 0.1353
IC– 17	7.96 ± 0.24	3.60 ± 0.11	8.70 ± 0.36	22.10 ± 0.88	0.0096 ± 0.1363
IC– 18	7.79 ± 0.18	3.37 ± 0.08	8.43 ± 0.37	23.13 ± 0.87	− 0.0043 ± 0.1345
IC– 19	8.03 ± 0.19	3.53 ± 0.08	8.76 ± 0.37	22.76 ± 0.88	0.0016 ± 0.1504
IC– 20	7.99 ± 0.25	3.58 ± 0.11	9.03 ± 0.38	22.32 ± 0.87	0.0017 ± 0.1495
IC– 21	7.62 ± 0.15	3.45 ± 0.07	8.23 ± 0.36	21.84 ± 0.88	0.0118 ± 0.1512
IC– 22	8.25 ± 0.26	3.56 ± 0.11	8.94 ± 0.37	23.19 ± 0.89	0.0048 ± 0.1370
IC– 23	7.97 ± 0.17	3.31 ± 0.06	8.65 ± 0.37	24.28 ± 0.76	− 0.0007 ± 0.1372
IC– 24	8.22 ± 0.29	3.58 ± 0.13	9.15 ± 0.38	22.95 ± 0.88	0.0017 ± 0.1358
IC– 25	8.14 ± 0.23	3.39 ± 0.10	10.2 ± 0.39	24.01 ± 0.88	− 0.0045 ± 0.1361
IC– 26	7.80 ± 0.14	3.48 ± 0.06	8.83 ± 0.38	22.41 ± 0.87	− 0.0013 ± 0.1339
IC– 27	7.60 ± 0.22	3.54 ± 0.10	9.81 ± 1.80	21.50 ± 0.87	0.0066 ± 0.1366
IC– 28	7.87 ± 0.11	3.47 ± 0.04	8.63 ± 0.37	22.68 ± 0.87	− 0.0018 ± 0.1343
IC– 29	7.69 ± 0.89	3.51 ± 0.04	8.54 ± 0.37	21.93 ± 0.87	0.0012 ± 0.1337
IC-30	7.76 ± 0.21	3.27 ± 0.09	8.69 ± 0.37	23.75 ± 0.87	− 0.0081 ± 0.1346
IC-31	7.73 ± 0.18	3.39 ± 0.08	8.55 ± 0.37	22.76 ± 0.88	− 0.0002 ± 0.1349
IC-32	7.85 ± 0.18	3.33 ± 0.08	8.86 ± 0.38	23.60 ± 1.11	− 0.0037 ± 0.1901
IC-33	7.99 ± 0.32	3.60 ± 0.14	9.33 ± 0.39	22.19 ± 0.87	− 0.0001 ± 0.1339
IC-34	7.70 ± 0.95	3.42 ± 0.04	8.86 ± 0.37	22.53 ± 0.29	− 0.0019 ± 0.1340
IC-35	7.81 ± 0.50	3.56 ± 0.23	8.70 ± 0.37	21.94 ± 0.07	0.0149 ± 0.1410
IC-37	7.87 ± 0.12	3.18 ± 0.05	8.11 ± 0.35	24.79 ± 0.90	− 0.0012 ± 0.1383
IC-38	7.55 ± 0.14	3.07 ± 0.06	7.70 ± 0.33	24.57 ± 0.91	0.0048 ± 0.1396
IC-39	7.61 ± 0.25	2.97 ± 0.10	7.86 ± 0.34	25.59 ± 0.91	− 0.0020 ± 0.1395
IC-40	7.14 ± 0.17	2.93 ± 0.07	7.17 ± 0.33	24.39 ± 0.90	0.0026 ± 0.1387
AVC***	7.82 ± 0.21	3.43 ± 0.21	8.72 ± 0.55	22.81 ± 1.19	0.0009 ± 0.0069
FPTT1$^{\&}$	17.5	8.89	66.2	19.29	−0.0304
FPTT2	2.49 ± 0.07	0.36 ± 0.01	0.60 ± 0.05	68.71 ± 1.26	0.0435 ± 0.1940
CT	31.4 ± 0.2	23.9 ± 1.0	248 ± 1	13.15 ± 0.62	− 0.0522 ± 0.0962

*average and standard deviation of the set of monitors ^{198}Au, ^{59}Fe, ^{69m}Zn, ^{65}Zn and ^{95}Zr

**uncertainty with k = 1 calculated from $^{54}Fe(n,p)^{54}Mn$ at 834.8 keV (major contribution from σ_f = (77.016 ± 2.707) mb [28], net peak area and ε_p)

***average and standard deviation of all data in the carousel

$^{\#}$, Not included in average calculations

$^{+}$, Determined in 2018

$^{++}$ Determined in 2019 [17]; FPTT2, Fast Transfer Pneumatic Terminal 2; CT, Central Thimble

$^{\&}$, Determined in F12 in 2005

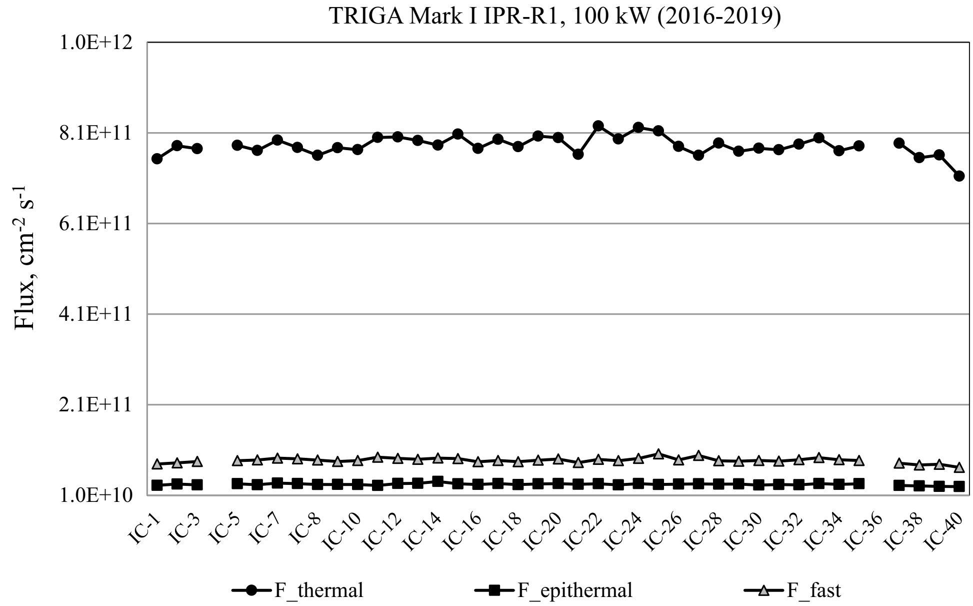

Fig. 2 Distribution of neutron fluxes in the irradiation channels of Carousel (IC-4 and IC-36 were unable)

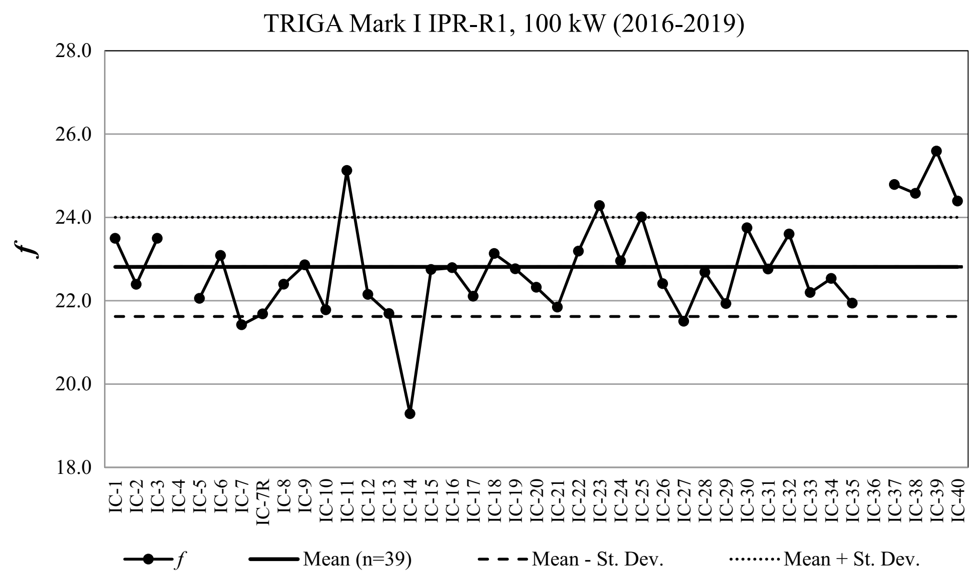

Fig. 3 Distribution of spectral parameter *f* in the irradiation channels of Carousel (IC-4 and IC-36 were unable)

determined in 2018 and 2019 (IC-7 and IC-7R) [17] and also determined at the end of 2021 (IC-721), after the pandemic period due to SARS-CoV− 2 virus causing COVID− 19 disease. During this time, the TRIGA IPR-R1 reactor was out of operation. The objective was to verify if the values were still similar to those measured before the long period without operation.

In the Carousel, it was not possible to determine the values in two irradiation channels (IC-4 and IC-36) because they were unavailable. In order to have one overview about values of spectral parameters and associated standard deviations in the carousel facility of the TRIGA reactor their averaged values for Carousel (AVC) were calculated. To calculate the AVC all values determined for all irradiation channels were included except values after the period for IC-721. The reason is to compare results obtained before and after period when the reactor was out of operation. For IC− 27, the fast neutron flux presented an unexpected high uncertainty (see Table 3). The experimental procedure to determine the spectral parameters and neutron fluxes should be repeated afterwards.

Table 3 shows the values for each irradiation channel in Carousel, in Fast Pneumatic Transfer Terminals 1 (FPTT1) and 2 (FPTT2) and in Central Thimble (CT) including the AVC. As can be seen, the AVC for thermal, epithermal and fast fluxes deviation is about 2.7%, 6.1% and 6.3%,

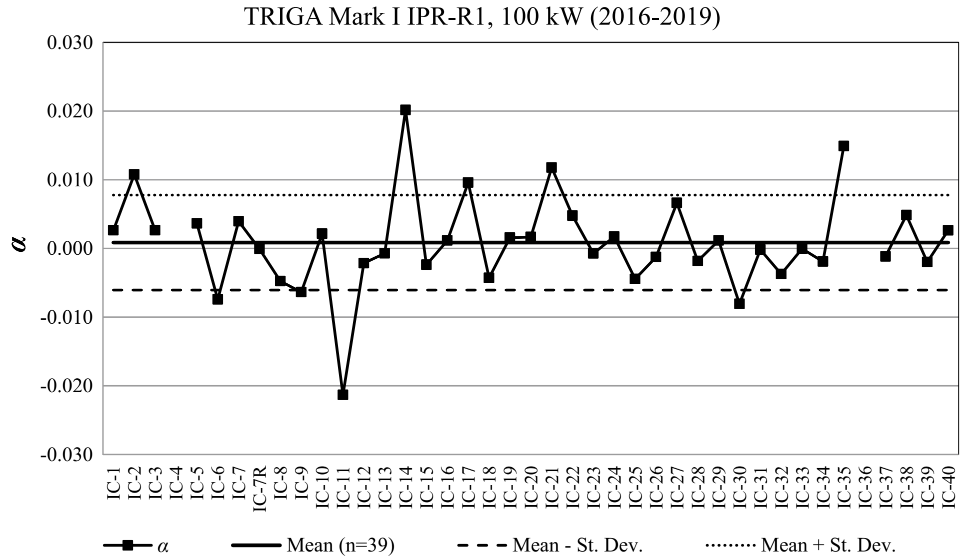

Fig. 4 Distribution of spectral parameter α *i*n the irradiation channels of Carousel (IC-4 and IC-36 were unable)

Table 4 Mass fraction of elements in BCR-320R irradiated in IC-7R, 2019, and IC− 11 and the percentage difference between the referential (AVC) and specific channel

El.	IC-7R (mg/kg)	Referential Values IC-7$_{AVC}$ (mg/kg)	PD %	IC− 11 (mg/kg)	Referential Values IC− 11$_{AVC}$ (mg/kg)	PD %
As	21.8 ± 0.8	21.6 ± 0.8	0.7	23.9 ± 0.8	24.2 ± 0.9	− 1.6
Au	0.009 ± 0.001	0.009 ± 0.001	−3.7	0.009 ± 0.001	0.009 ± 0.001	0.1
Ba	275 ± 17	270 ± 15	1.9	308 ± 14	316 ± 15	− 2.4
Br	75.8 ± 2.7	75.4 ± 2.6	0.5	85.8 ± 2.9	85.3 ± 3.0	− 2.9
Ca	35,560 ± 2146	34,430 ± 2078	3.3	39,860 ± 1914	39,480 ± 2191	1.0
Ce	33.8 ± 1.2	33.2 ± 1.2	2.0	36.7 ± 1.3	37.5 ± 1.3	− 2.3
Co	9.44 ± 0.34	9.28 ± 0.33	1.7	10.3 ± 0.4	10.6 ± 0.4	− 2.8
Cr	58.1 ± 2.1	56.8 ± 2.0	2.2	63.7 ± 2.3	65.2 ± 2.4	− 2.2
Cs	4.18 ± 0.15	4.17 ± 0.15	0.3	4.63 ± 0.16	4.67 ± 0.17	− 0.7
Eu	0.650 ± 0.048	0.642 ± 0.047	1.1	0.706 ± 0.051	0.717 ± 0.052	− 1.2
Fe	24,730 ± 869	24,260 ± 853	1.9	26,750 ± 943	27,380 ± 966	− 2.3
Hf	4.66 ± 0.17	4.59 ± 0.16	1.6	5.12 ± 0.18	5.24 ± 0.19	− 2.3
K	12,160 ± 456	11,950 ± 446	1.8	12,790 ± 478	13,090 ± 490	− 2.3
La	19.0 ± 0.7	18.5 ± 0.6	2.8	19.5 ± 0.8	21.2 ± 0.7	− 8.1
Na	9045 ± 321	8840 ± 310	2.3	9901 ± 347	10,130 ± 355	− 2.3
Nd	17.2 ± 0.7	16.9 ± 0.7	1.7	19.3 ± 0.7	19.9 ± 0.8	− 2.5
Rb	60.3 ± 2.3	60.3 ± 2.3	− 0.1	66.0 ± 2.4	69.1 ± 2.5	-4.2
Sb	1.01 ± 0.04	1.02 ± 0.04	− 0.9	1.18 ± 0.05	1.19 ± 0.05	− 1.0
Sc	5.14 ± 0.18	5.04 ± 0.18	2.1	5.59 ± 0.20	5.72 ± 0.20	− 2.2
Sm	2.80 ± 0.11	2.77 ± 0.12	1.0	3.11 ± 0.11	3.12 ± 0.11	− 0.4
Sr	186 ± 12	186 ± 12	0.1	199 ± 12	206 ± 13	-3.6
Ta	0.451 ± 0.018	0.456 ± 0.018	− 1.0	0.527 ± 0.021	0.525 ± 0.021	0.3
Tb	0.392 ± 0.015	0.393 ± 0.015	− 0.2	0.431 ± 0.016	0.435 ± 0.016	− 0.9
Th	4.89 ± 0.17	4.88 ± 0.17	0.3	5.34 ± 0.19	5.45 ± 0.19	− 2.0
U	1.44 ± 0.18	1.40 ± 0.07	3.1	1.62 ± 0.08	1.64 ± 0.07	− 1.0
Yb	1.42 ± 0.06	1.37 ± 0.06	3.5	1.49 ± 0.07	1.52 ± 0.20	− 2.2
Zn	311 ± 11	306 ± 11	1.7	342 ± 12	352 ± 13	− 2.7
Zr	183 ± 12	181 ± 12	1.0	206 ± 13	215 ± 13	− 4.0

El. Element; *, Calculated with data of IC-7R, 2019 (see Table 3); PD, Percentage Difference

Table 5 Mass fraction of elements in BCR-320R irradiated in IC− 14 and IC− 20 and the percentage difference between the referential (AVC) and specific channel

El.	IC− 14 (mg/kg)	Referential Values IC− 14_{AVC} (mg/kg)	PD %	IC− 20 (mg/kg)	Referential Values IC− 20_{AVC} (mg/kg)	PD %
As	23.6 ± 15	23.9 ± 0.9	1.4	22.4 ± 0.8	22.4 ± 0.8	0.1
Au	0.009 ± 0.001	0.009 ± 0.001	4.5	0.010 ± 0.001	0.010 ± 0.001	0.0
Ba	305 ± 14	287 ± 13	6.2	287 ± 13	287 ± 13	− 0.1
Br	88.8 ± 3.1	83.0 ± 2.9	7.0	77.4 ± 2.7	77.5 ± 2.7	0.0
Ca	41,960 ± 2327	38,270 ± 1591	9.6	33,340 ± 2352	33,110 ± 2336	0.7
Ce	40.0 ± 1.4	36.2 ± 1.3	10.4	34.0 ± 1.2	33.7 ± 1.2	0.7
Co	10.9 ± 0.6	9.9 ± 0.4	9.8	9.40 ± 0.34	9.34 ± 0.33	0.6
Cr	67.1 ± 3.7	60.8 ± 2.2	10.3	57.7 ± 2.0	57.3 ± 2.0	0.8
Cs	4.71 ± 0.17	4.46 ± 0.16	5.7	4.20 ± 0.15	4.20 ± 0.15	0.1
Eu	0.700 ± 0.048	0.651 ± 0.045	7.6	0.629 ± 0.044	0.626 ± 0.044	0.4
Fe	25,620 ± 1142	25,890 ± 908	5.5	24,550 ± 861	24,380 ± 855	0.7
Hf	5.43 ± 0.20	4.95 ± 0.19	9.6	4.61 ± 0.16	4.58 ± 0.17	0.7
K	13,920 ± 525	12,670 ± 473	9.9	12,070 ± 453	11,980 ± 450	0.8
La	22.6 ± 0.8	20.6 ± 0.7	10.1	19.2 ± 0.7	19.1 ± 0.7	0.7
Na	10,610 ± 373	9615 ± 338	10.3	9181 ± 322	9031 ± 320	1.7
Nd	20.6 ± 1.2	18.7 ± 1.1	10.2	17.6 ± 1.1	17.5 ± 1.1	0.7
Rb	71.4 ± 2.7	65.6 ± 2.4	8.9	59.7 ± 2.2	59.6 ± 2.2	0.2
Sb	1.23 ± 0.05	1.18 ± 0.05	4.3	1.08 ± 0.04	1.09 ± 0.04	− 0.3
Sc	5.41 ± 0.27	5.37 ± 0.19	0.6	5.07 ± 0.18	5.03 ± 0.18	0.8
Sm	3.24 ± 0.12	3.09 ± 0.11	5.0	2.85 ± 0.11	2.69 ± 0.12	6.2
Sr	215 ± 13	198 ± 12	8.5	193 ± 13	193 ± 14	0.2
Ta	0.530 ± 0.021	0.516 ± 0.020	2.7	0.438 ± 0.017	0.434 ± 0.018	1.0
Tb	0.441 ± 0.017	0.419 ± 0.016	5.2	0.391 ± 0.015	0.391 ± 0.015	− 0.1
Th	5.89 ± 0.22	5.49 ± 0.20	7.2	5.01 ± 0.18	5.00 ± 0.18	0.2
U	1.62 ± 0.07	1.56 ± 0.07	4.1	1.42 ± 0.06	1.43 ± 0.06	− 0.7
Yb	1.64 ± 0.08	1.47 ± 0.06	11.5	1.41 ± 0.07	1.40 ± 0.07	0.7
Zn	366 ± 22	332 ± 12	10.3	317 ± 11	314 ± 11	0.7
Zr	218 ± 16	196 ± 14	10.9	169 ± 12	167 ± 10	1.4

El. Element; PD, Percentage Difference

respectively, for f is about 5.2%, while for the parameter α the deviation is about 770% due to the very low values. The Fast Pneumatic Transfer Terminal 1, to be installed in F12 in the core, will be used to produce radionuclides with short half-life and the elemental concentrations will be analysed by k_0-method. The spectral parameters and neutron fluxes were determined in 2005 using the bare method [18] with ^{198}Au– ^{95}Zr monitors. It was a preliminary assessment about the values in this channel aiming at future installation of pneumatic transfer for short half-live radionuclides analysis. The spectral parameters and neutron flux will be determined again more precisely to have them available. FPTT2 and Central Thimble are not used to apply the k_0-method, but it is interesting to be available for other studies.

The values for IC-721 were not included in the figures. Figure 2 shows the distribution of neutron fluxes in the Carousel and Figs. 3 and 4 display the distribution of spectral parameters f and α.

Related to the Carousel, it can be observed that the irradiation channels IC− 11 and IC-39, presented the highest values for f and IC− 14, the lowest value. In order to verify if these data would interfere in the calculations, aliquots of certified reference material BCR-320R were irradiated in these channels. This material was also irradiated in channel IC-7 (as irradiation channel reference [13]) and in IC− 20 (chosen by chance).

To calculate the mass fractions of the aliquots of the reference material by k_0-method, it was necessary to determine the $F_{c,Au}$ [17, 29]. For each sample, two values for $F_{c,Au}$ were calculated: one called $F_{c,Au\text{-}IC}$ using the spectral parameters values of the channel and other, $F_{c,Au\text{-}AVC}$, calculated using the average spectral parameters for Carousel. Therefore, the mass fractions were obtained using the values of f_{IC}, α_{IC} and $F_{c,Au\text{-}IC}$ [17, 29] of specific channel and using f_{AVC}, α_{AVC} and $F_{c\text{-}AVC}$ values of the Carousel. For IC-7, the values used correspond to IC-7R determined in 2019, the last value before the reactor was out of operation. The results determined for

Table 6 Mass fraction of elements in BCR-320R irradiated in IC-39 and the percentage difference between the referential (AVC) and specific channel

El.	IC-39 (mg/kg)	Referential Values $IC\text{-}39_{AVC}$ (mg/kg)	PD %
As	22.4 ± 0.8	22.7 ± 0.8	− 1.6
Au	0.010 ± 0.001	0.011 ± 0.001	− 1.4
Ba	270 ± 12	248 ± 10	8.8
Br	78 ± 3	79 ± 3	− 1.1
Ca	34,900 ± 1904	36,680 ± 2154	− 4.9
Ce	34.9 ± 1.2	36.4 ± 1.3	− 4.2
Co	9.67 ± 0.34	10.0 ± 0.4	− 3.5
Cr	59.6 ± 2.1	62.8 ± 2.2	− 5.0
Cs	4.31 ± 0.15	4.21 ± 0.15	2.3
Eu	0.648 ± 0.048	0.705 ± 0.045	− 8.2
Fe	24,940 ± 876	25,980 ± 911	-4.0
Hf	4.80 ± 0.21	4.97 ± 0.21	− 8.8
K	12,040 ± 461	12,450 ± 473	− 3.3
La	19.3 ± 0.7	20.3 ± 0.7	− 4.8
Na	9126 ± 320	9523 ± 335	− 4.2
Nd	18.8 ± 0.9	19.5 ± 0.9	− 3.8
Rb	61.7 ± 2.3	62.2 ± 2.4	− 0.9
Sb	1.10 ± 0.04	1.09 ± 0.04	0.6
Sc	5.19 ± 0.18	5.44 ± 0.20	− 4.7
Sm	2.93 ± 0.11	2.97 ± 0.11	− 1.4
Sr	195 ± 14	197 ± 14	− 1.1
Ta	0.466 ± 0.019	0.459 ± 0.019	1.7
Tb	0.408 ± 0.016	0.410 ± 0.016	− 0.5
Th	5.16 ± 0.18	5.22 ± 0.20	− 1.3
U	1.54 ± 0.06	1.50 ± 0.06	2.5
Yb	1.42 ± 0.07	1.49 ± 0.06	− 4.9
Zn	318 ± 11	331 ± 12	− 3.8
Zr	208 ± 14	219 ± 29	− 5.4

El. Element; PD, Percentage Difference

Table 7 Values of f, α and F_C used for mass fractions calculations

IC	f_{IC}	α_{IC}	$F_{c,Au\text{-}IC}$	$F_{c,Au\text{-}AVC}$
7 (2018)	21.42	0.0039	10,872	10,459
7R (2019)	21.68	− 0.0001	10,768	
721 (2021)	23.25	− 0.0045	10,303	
11	25.12	− 0.0213	9869	10,618
14	19.29	0.0201	11,042	10,333
20	22.32	0.0017	10,316	10,175
39	25.59	− 0.0020	8778	9431

IC, Irradiation Channel; f_{IC}, Value of parameter f for specific IC; α_{IC}, Value of parameter α for specific IC; $F_{c,Au\text{-}IC}$, Comparator Factor for gold monitor calculated for specific IC; $F_{c,Au\text{-}AVC}$, Average of Comparator Factor for gold monitor calculated for all Irradiation Channels in the Carousel

all elements are displayed in Table 4, for IC-7R and IC− 11; Table 5, for IC− 14 and IC− 20 and Table 6 for IC-39. The software Kayzero for Windows®, V3.37 [20] was applied for mass fraction calculations. Table 7 displays all values of f, α and $F_{c,Au}$ used for the calculations.

Tables 4, 5 and 6 also display the percentage difference, PD. It was calculated in order to determine how close were the results obtained based on values of each channel and the values of the average carousel considered the referential values. PD is an interesting evaluator that points out if it is possible to calculate the mass fractions based on IC parameters or Carousel average parameters values.

PD shows that all percentage differences are lower than 10% for IC-7R and its related $IC\text{-}7R_{AVC}$, IC− 11 and IC− 11_{AVC}, IC− 20 and IC− 20_{AVC} as well as IC-39 and $IC\text{-}39_{AVC}$. For IC− 14 and IC− 14_{AVC}, the values lower than 10% correspond to 71.4% however, the maximum difference is lower than 12%. It is interesting that the value for parameter f in IC− 14 is the lowest among all values for f determined.

Therefore, evaluating the results related to certified results by E_n-score, Table 8, all values are $|E_n| \leq 1.0$, pointing out that the methodology applied produced results with 95% probability to be inside a range of values that correspond to the certified values.

The evaluator E_n-score was also applied to BCR-320R sample irradiated in IC-7, using the spectral parameters and fluxes determined for this channel in 2018 (IC-7), 2019 (IC-7R) and in 2021 (IC-721). Table 9 shows the E_n-score results.

Conclusion

The values for thermal, epithermal and fast neutron fluxes and f and α spectral parameters were determined by the most accurate procedure, "Cd-ratio for multi-monitor" method in all irradiation devices of TRIGA Mark I IPR-R1 reactor – Carousel, Fast Pneumatic Transfer System 2 and Central Thimble. For the first time, the values determined in all irradiation devices were based on the same procedure and using the same materials (certified monitors and neutron filter).

The objective was to enable the analysis by the k_0-standardized method of neutron activation analysis in the irradiation channels in the Carousel, aiming at producing reliable elemental concentration values. The values were also determined in the Fast Pneumatic Transfer System 2 and Central Thimble as additional information.

The test E_n-score was applied to experimental values of the aliquots of reference materials BCR-320R irradiated in specific irradiation channels. The channels were chosen based on the highest and the lowest found values of f. Additionally a reference channel was chosen as well as a random

Table 8 Certified values for BCR-320R [25] and experimental results evaluated by E_n-score. BCR-320R samples were irradiated in IC-7, IC− 11, IC− 14, IC− 20 and IC-39 and the calculations were based on specific irradiation channel (IC-X $_{f,\alpha}$, X = 7, 11, 14, 20, 39) and on Carousel average values (AVC$_{f,\alpha}$)

BCR-320R

El.	Certified Values (mg/kg) $k=2$	E_n-score IC-7R$_{f,\alpha}$	AVC$_{f,\alpha}$	IC− 11$_{f,\alpha}$	AVC$_{f,\alpha}$	IC− 14$_{f,\alpha}$	AVC$_{f,\alpha}$	IC− 20$_{f,\alpha}$	AVC$_{f,\alpha}$	IC-39$_{f,\alpha}$	AVC$_{f,\alpha}$
As	21.7 ± 2.0	0.03	− 0.04	0.82	0.94	0.51	0.83	0.28	0.27	0.25	0.40
Co	9.7 ± 0.6	− 0.29	− 0.47	0.62	0.93	0.88	0.20	− 0.34	− 0.41	− 0.04	0.34
Cr	59 ± 4	− 0.16	− 0.38	0.78	0.96	0.97	0.31	− 0.23	− 0.31	0.10	0.63
Fe	25,700 ± 1300	− 0.45	− 0.67	0.46	0.72	− 0.03	0.09	− 0.53	− 0.61	− 0.35	0.13
Sc	5.2 ± 0.4	− 0.10	− 0.30	0.70	0.91	0.31	0.31	− 0.24	− 0.31	− 0.03	0.43
Th	5.3 ± 0.4	− 0.77	− 0.80	0.07	0.26	0.99	0.34	− 0.54	− 0.56	− 0.27	− 0.13
U	1.56 ± 0.20	− 0.30	− 0.67	0.24	0.33	0.26	0.00	− 0.60	− 0.56	− 0.10	− 0.26
Zn	319 ± 20	− 0.27	− 0.45	0.74	0.98	0.98	0.41	− 0.08	− 0.15	− 0.03	0.37

El., Element; IC, Irradiation Channel; AVC, Average Value for Carousel

channel. Comparing the values of mass fractions calculated with spectral parameters and fluxes for that channel and the average values for Carousel, the highest percentage difference was about 12%. However, applying the statistical test E_n-score, the results pointed out that the k_0-method is producing reliable results for a specific channel or using the Carousel average parameters.

The mass fractions of reference sample irradiated in the IC-7, our reference irradiation channel [13], were calculated using data (spectral parameters and neutron fluxes) obtained in 2028, 2019 and 2021 for IC-7. The statistical test E_n-score also pointed out that along the time the k_0-method is producing results with 95% of possibility to be inside a range of values that correspond to the certified values. They also showed that possible changes in the reactor due to the period that it was out of operation are inside the uncertainty.

Table 9 Certified values for BCR-320R [25] and experimental results evaluated by E_n-score. BCR-320R sample was irradiated in IC-7 and the calculations were based on spectral values and fluxes determined in 2018 (IC-7), 2019 (IC-7R) and 2021 (IC-721), Table 3

BCR-320R

El.	Certified Values [25] (mg/kg) $k=2$	E_n-score IC-7 (2018)	IC-7R (2019)	IC-721 (2021)
As	21.7 ± 2.0	0.01	0.03	− 0.22
Co	9.7 ± 0.6	− 0.26	− 0.29	− 0.67
Cr	59 ± 4	− 0.14	− 0.16	− 0.57
Fe	25,700 ± 1300	− 0.41	− 0.45	− 0.90
Sc	5.2 ± 0.4	− 0.08	− 0.10	− 0.48
Th	5.3 ± 0.4	− 0.74	− 0.77	− 0.98
U	1.56 ± 0.2	− 0.77	− 0.30	− 0.76
Zn	319 ± 20	− 0.22	− 0.27	− 0.66

From now on all calculations related to determination of mass fractions by k_0-INAA method as well as production and studies of radioisotopes (radiopharmaceuticals, radiotracers and production of radioactive sources) will be more accurate including their calculated induced activities.

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