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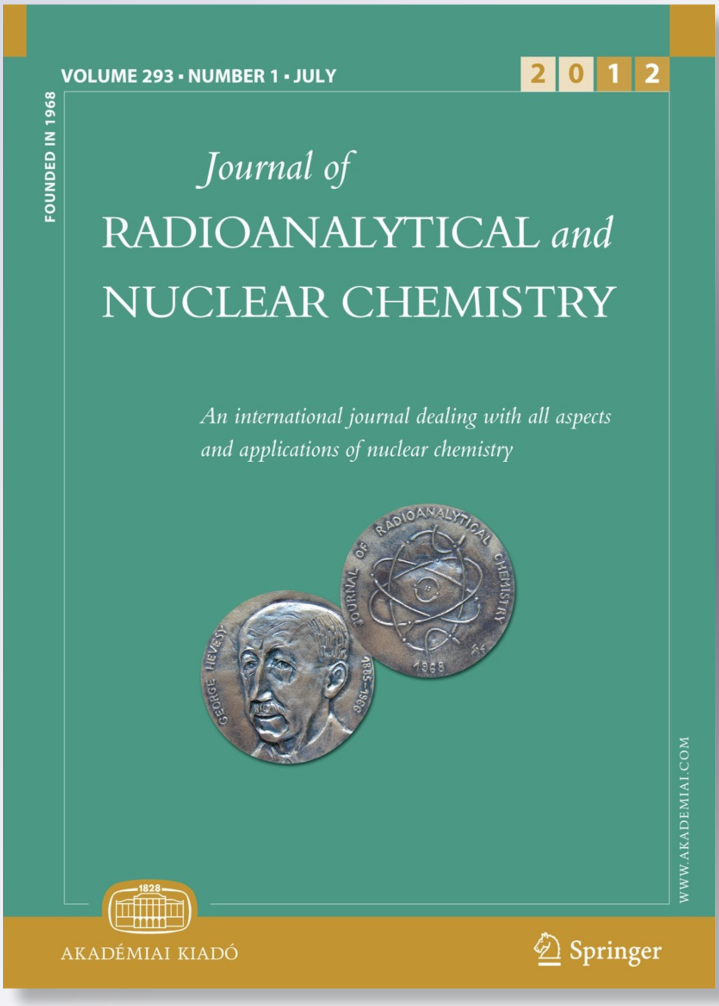

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Abstract We focus on identifying quantitatively the effects on activation measurements due to temporal variations of the neutron flux. Irradiations in the carousel facility (CF) of the TRIGA reactor at the Jožef Stefan Institute (JSI) for core No. 176 (April 2002) and current core No. 189 (June 2006), are discussed for illustration. The flux measurements are based on neutron detectors (ionisation chambers), outside the graphite reflector. The variations of the neutron flux produce a systematic error in the results obtained by the neutron activation analysis by the k_0-standardization method, which assumes constant conditions during irradiation. The results of our study show that for a typical irradiation of 20 h in the channels of the CF, aligned in the direction of the ionisation chamber (safety channel), the temporal variation of the neutron flux is about 6−8%. In the k_0 method, which we are using for routine work at the JSI, this variation introduced a systematic error in the results after long irradiation of 20 h up to 5%, depending on the half-life of the investigated radionuclide.

Keywords Temporal variation · Neutron flux · Saturation factor · k_0-NAA

Introduction

Most analyses of samples carried out at the Department of Environmental Sciences of the JSI pertain to environmental monitoring surveys and health-related studies using the activation techniques in steady-state operation mode of the 250 kW TRIGA Mark II reactor (see Fig. 1). More specifically, to determine a relatively wide range of elements in different samples the k_0-standardization method of NAA is used. The k_0-method is a “quasi” absolute technique, which uses gold as the standard: composite nuclear constants for analytically interesting nuclides are normalised to the gold nuclear data (molar mass, isotopic abundance, cross-section, absolute γ-intensity). The method in its basic form assumes uninterrupted and constant irradiation conditions, which may not always be the case.

This work is focused on calculation of correction factors based on observed temporal variation of the neutron flux in the CF of TRIGA reactor for core No. 176 (April 2002) and current core No. 189 (as of June 2006), for long irradiation periods. The reactor core No. 189 differs from core No. 176 as follows: fresh fuel element was placed in position E19, neutron source was moved from position E7 to F8 and the E7 position was filled with a fresh fuel element. The influence of the variations in flux on the results obtained by the k_0-method for some analytically interesting nuclides are also presented.

Method

k_0-based method of NAA

The k_0-based standardization method assumes that spectral parameters f (thermal-to-epithermal flux ratio) and α (deviation from 1/E distribution of epithermal neutrons) are constant during irradiation and known. For a constant flux during irradiation the basic equation of the k_0-method [1, 2] is:

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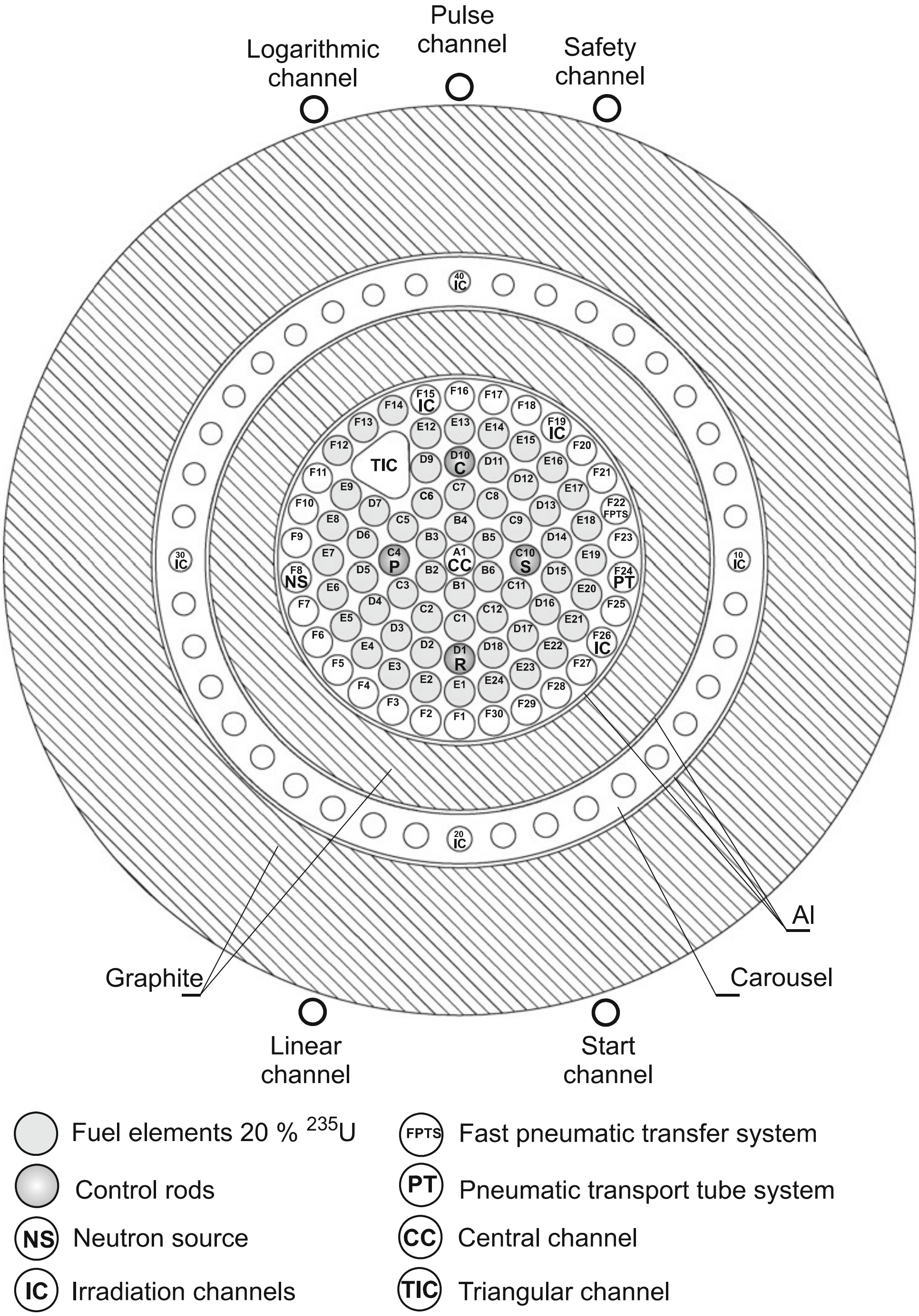

Fig. 1 Ground plan of the TRIGA Mark II reactor (core No. 189) with irradiation channels

$$\rho_a = \frac{\left(\frac{N_p/t_m}{SDCw}\right)_a}{\left(\frac{N_p/t_m}{SDCw}\right)_c} \frac{1}{k_{0,c}(a)} \frac{G_{th,c}f + G_{e,c}Q_{0,c}(\alpha)}{G_{th,a}f + G_{e,a}Q_{0,a}(\alpha)} \frac{\varepsilon_{p,c}}{\varepsilon_{p,a}} \qquad (1)$$

where index a refers to the investigated nuclide, index c to the comparator ^{198}Au ($T_{1/2} = 2.695$ days) and saturation factor S during irradiation expressed as $S = 1 - e^{-\lambda t_{\mathrm{irr}}}$.

When the neutron flux varies during irradiation and the parameters f and α remain constant, S in the expression (1) has to be replaced by:

$$S' = \int_0^{t_{\mathrm{irr}}} F(t)\lambda\, e^{\lambda(t - t_{\mathrm{irr}})}\, dt \qquad (2)$$

where $F(t)$ is a function of the time-dependent neutron flux, normalised so that it equals 1 zero-time irradiations. Correction of the saturation factor (S′) depends on the half-life and irradiation time. In a first approximation, linear time dependence of the neutron flux can be written as:

$$F(t) = 1 + k \cdot t \tag{3}$$

where k is the slope of neutron flux during irradiation. After substituting expression (3) into expression (2) and integrating with respect to time, we obtain:

$$S' = S\left(1 + k \cdot t_{\mathrm{irr}}\left(\frac{1}{S} - \frac{1}{\lambda \cdot t_{\mathrm{irr}}}\right)\right) \tag{4}$$

The impact of the corrected saturation factor on the final result (F_{sat}) is expressed as a deviation in percent of the measured nuclide concentration due to changes in neutron flux compared to the reference concentration at constant neutron flux:

$$F_{\mathrm{sat}} = \left(\frac{S'_c S_a}{S'_a S_c} - 1\right) \cdot 100 \tag{5}$$

From Eqs. 5 and 1 it can be seen that when $k < 0$ and the half-life of investigated radionuclide $(T_{1/2})_{\mathrm{a}}$ is less than the half-life of ^{198}Au then the final result should be increased ($F_{\mathrm{sat}} > 0$) and vice versa.

Steady state operation mode of the TRIGA reactor

The operators of the TRIGA reactor have five independent neutron detectors (start, linear, logarithmic, pulse and safety channels (Fig. 1)) to control the power of the reactor. To make the reactor critical, the operator fixes the position of the compensating rod (labelled C in Fig. 1), which is alligned radially in the direction of the IC-40 channel, where samples are usually irradiated. Reactivity change due to temperature and xenon buildup is compensated by the movement of the regulating control rod (labelled R), which causes spatial neutron flux

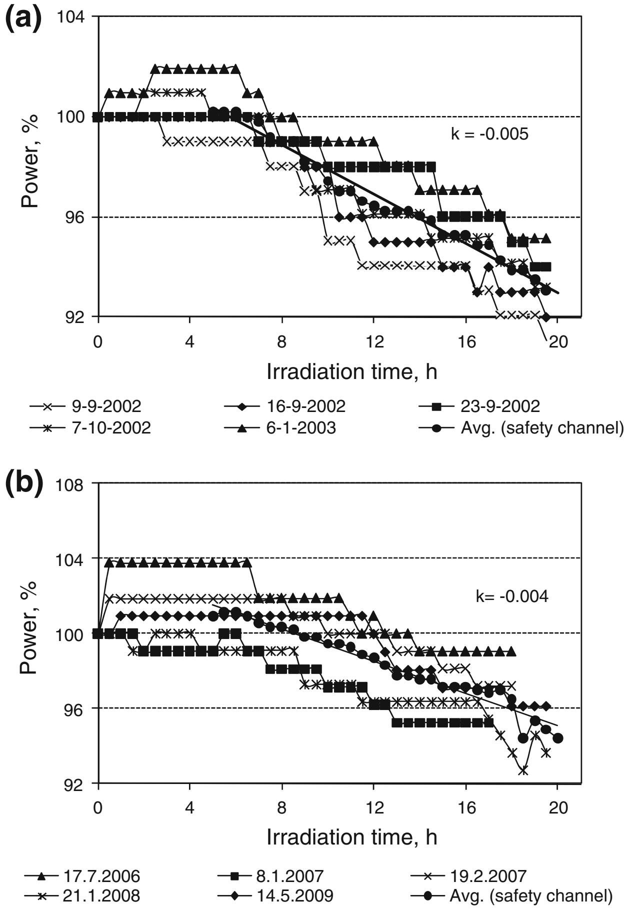

Fig. 2 Time-dependence of the neutron flux readings from the safety channel during uninterrupted irradiation of 20 h at "constant power level" as measured from the linear channel. **a** Reactor core set up No. 176. **b** Reactor core set up No. 189. Parameter k is the slope [5]

Table 1 Typical positions of compensating and regulating control rods during steady state operation of TRIGA reactor for two different core configurations

Irradiation	Position of compensating rod	Position of regulating rod	Slope [1/h] (from 5 to 20 h)
Core No. 176			
9–10 Sep. 2002	400	From 420 to 263	$k = -0.0055$
16–17 Sep. 2002	400	From 426 to 258	$k = -0.0059$
23–24 Sep. 2002	400	From 418 to 231	$k = -0.0039$
7–8 Oct. 2002	350	From 428 to 262	$k = -0.0050$
6–7 Jan. 2003	350	From 426 to 282	$k = -0.0046$
Core No. 189			
17–18 Jul. 2006	350	From 504 to 397	$k = -0.0041$
8–9 Jan. 2007	460	From 417 to 293	$k = -0.0043$
19–20 Feb. 2007	380	From 473 to 364	$k = -0.0041$
21–22 Jan. 2008	400	From 429 to 270	$k = -0.0038$
14–15 May 2009	400	From 405 to 215	$k = -0.0042$

redistribution [3]. The nominal power of 250 kW is kept constant by the control system connected to the signal from the linear channel. The power from the linear and logarithmic channels is recorded on strip-chart paper; the power from the pulse and the safety channels is read off visually and recorded into the operators' logbook. Other relevant information, such as temperature of the coolant (water) and the fuel elements, are also recorded. This information is used to quantify the flux variation in different irradiation channels in the CF and to determine the correction to the saturation factor in the standard procedure of the k_0-method of NAA.

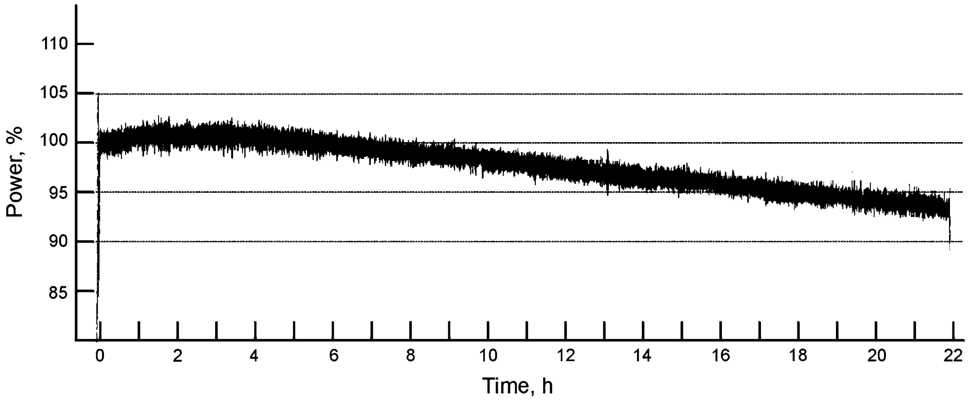

Fig. 3 The reactor power for core No. 176 reading with electrometer from the safety channel from 27 to 28 Jan 2003 [5, 6]

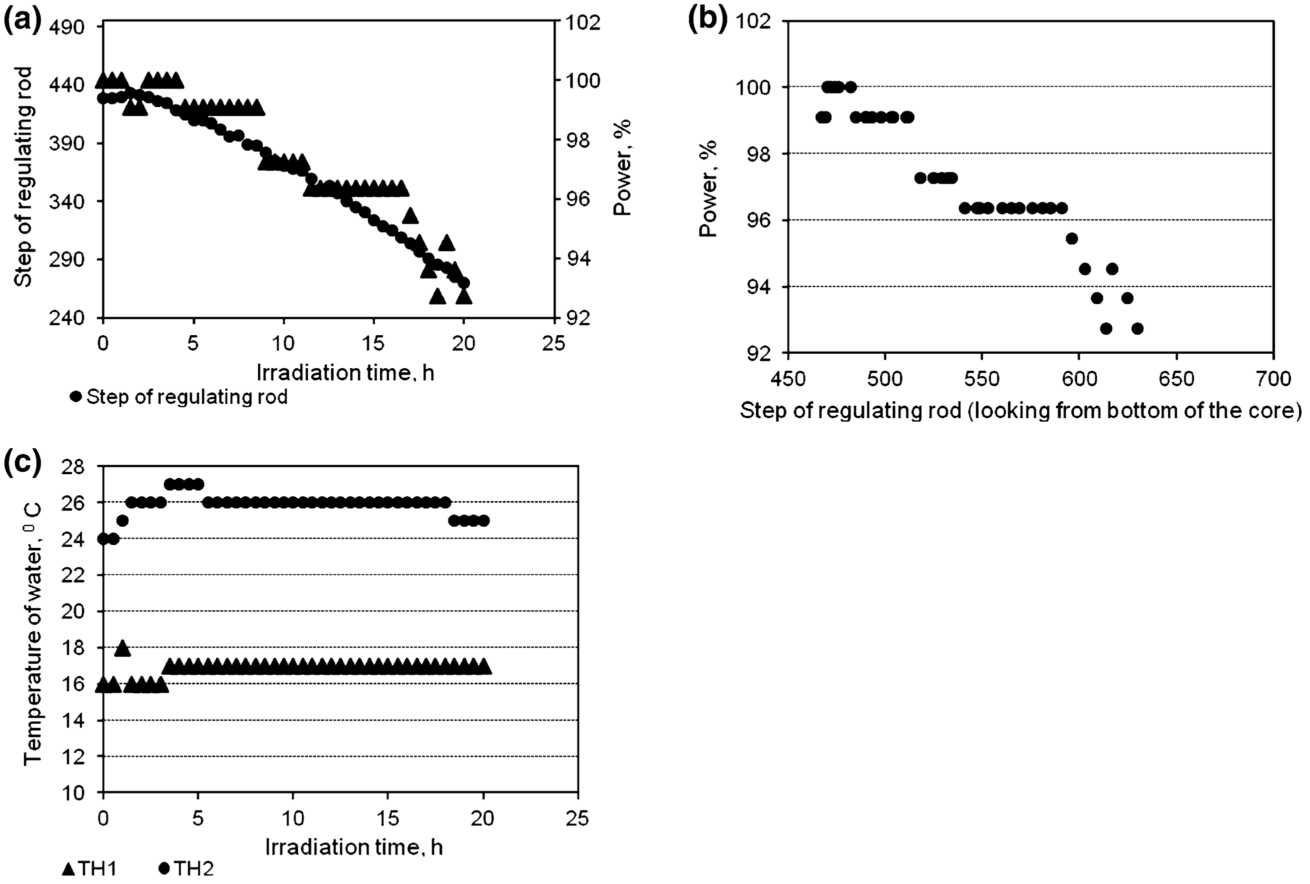

Fig. 4 Reactor power for core set up No. 189 as measured from the safety channel and regulating rod positions during irradiation of 20 h started at 21.1.2008 (**a**), correlation between the power and regulating rod position (**b**), temperature of coolant (water) during irradiation measured at the entrance near the reactor wall at 1 m depth, TH1 and at the mean of the pool water at 0.5 m depth, TH2 (**c**)

Table 2 Correction factors F_{sat} (in %) on the final result obtained by k_0-method for some nuclides originated from the changes of the neutron flux during long irradiations up to 20 h in the carousel facility at IC-40 channel

Element	Target isotope	Formed isotope	$T^{a}_{1/2}$ [8]	Activ. type[b] [4]	F_{sat} (%)			
					5 h	10 h	15 h	20 h
Au	^{197}Au	^{198}Au	2.695 d	I	0.00	0.00	0.00	0.00
Na	^{23}Na	^{24}Na	14.96 h	IVb	0.04	0.15	0.34	0.62
K	^{41}K	^{42}K	12.36 h	I	0.05	0.19	0.44	0.78
Ca	^{46}Ca	^{47}Ca	4.536 d	I	0.00	−0.02	−0.04	−0.08
		↓						
		^{47}Sc	3.349 d	IIa	−0.43	−0.87	−1.34	−1.82
	^{48}Ca	^{49}Ca	8.718 min	I	1.16	2.47	3.83	5.23
Mn	^{55}Mn	^{56}Mn	2.579 d	I	0.26	0.99	2.04	3.28
Fe	^{58}Fe	^{59}Fe	44.50 d	I	−0.01	−0.04	−0.10	−0.18
Co	^{59}Co	^{60m}Co	10.47 min	I	1.14	2.45	3.80	5.21
		↓						
		^{60}Co	5.271 y	IVb	−0.01	−0.05	−0.10	−0.19
Zn	^{64}Zn	^{65}Zn	244.3 d	I	−0.01	−0.05	−0.10	−0.19
	^{68}Zn	^{69m}Zn	13.76 h	I	0.04	0.17	0.38	0.69
	^{70}Zn	^{71}Zn	2.45 min	I	1.24	2.55	3.91	5.32
Mo	^{98}Mo	^{99}Mo	65.94 h	I	0.00	0.00	0.00	0.00
		↓						
		^{99m}Tc	6.01 h	IId	−0.30	−0.62	−0.91	−1.15
	^{100}Mo	^{101}Mo	14.61 min	I	1.09	2.40	3.75	5.15
		↓						
		^{101}Tc	14.2 min	IIa	0.91	2.21	3.56	4.95
Zr	^{94}Zr	^{95}Zr	60.02 d	I	−0.01	−0.04	−0.10	−0.18
		↓						
		^{95m}Nb	86.6 h					
		↓						
		^{95}Nb	34.97 d	IIIa	−0.43	−0.89	−1.38	−1.90
	^{96}Zr	^{97}Zr	16.74 h	I	0.03	0.13	0.30	0.53
		↓						
		^{97m}Nb	52.7 s	IIa	0.03	0.12	0.29	0.53
		↓						
		^{97}Nb	72.1 min	IIIa	−0.25	−0.28	−0.19	−0.01
Eu	^{151}Eu	^{152m}Eu	9.312 h	I	0.07	0.27	0.61	1.09
		No I.T.						
		^{152}Eu	13.54 y	I	−0.01	−0.05	−0.10	−0.19
	^{153}Eu	^{154}Eu	8.593 y	IVb	−0.01	−0.05	−0.10	−0.19
U	^{238}U	^{239}U	23.45 min	I	0.98	2.28	3.63	5.03
		↓						
		^{239}Np	2.357 d	IIb	0.00	0.01	0.01	0.03

The slope $k = -0.005$ is assumed in the calculation of the F_{sat}

[a] Half-life is taken from [8]

[b] For the activation-decay type classification, see [4]

Results and discussion

Due to the accumulation of xenon and partly due to temperature changes of the pool water, the core reactivity drifts and has to be compensated by control rod movement, particularly during long irradiation periods, lasting 20 h or more. Flux readings from several detectors around the core are available in the operators' logbook. Some typical

Table 3 Reduction of the correction factors F_{sat} (in %) on the final result obtained by k_0-method for some nuclides for chosen cooling (T_d) and measurement (T_m) time

Element	Nuclide	Activ. type [4]	T_d (h)	T_m (h)	F_{sat} (%)			
					5 h	10 h	15 h	20 h
Au	^{198}Au	I	0	0	0.00	0.00	0.00	0.00
			6.468[a]	6.468[a]	0.00	0.00	0.00	0.00
			64.68[b]	64.68[b]	0.00	0.00	0.00	0.00
			323.4[c]	64.68[b]	0.00	0.00	0.00	0.00
			646.8[d]	129.36[f]	0.00	0.00	0.00	0.00
			970.2[e]	194.04[g]	0.00	0.00	0.00	0.00
Na	^{24}Na	IVb	0	0	0.04	0.15	0.34	0.62
			1.496[a]	1.496[a]	0.04	0.15	0.34	0.62
			14.96[b]	14.96[b]	0.04	0.15	0.34	0.62
			74.8[c]	14.96[b]	0.04	0.15	0.34	0.62
			149.6[d]	29.92[f]	0.04	0.15	0.34	0.62
			224.4[e]	44.88[g]	0.04	0.15	0.34	0.62
Ca	^{47}Sc	IIa	0	0	−0.43	−0.87	−1.34	−1.82
			10.886[a]	10.886[a]	−0.06	−0.22	−0.44	−0.73
			108.86[b]	108.86[b]	−0.01	−0.04	−0.09	−0.16
			544.32[c]	108.86[b]	−0.01	−0.02	−0.05	−0.09
			1088.6[d]	217.73[f]	0.00	−0.02	−0.04	−0.08
			1633[e]	326.59[g]	0.00	−0.02	−0.04	−0.08
Mn	^{56}Mn	I	0	0	0.26	0.99	2.04	3.28
			0.2579[a]	0.2579[a]	0.26	0.99	2.04	3.28
			2.579[b]	2.579[b]	0.26	0.99	2.04	3.28
			12.895[c]	2.579[b]	0.26	0.99	2.04	3.28
			25.79[d]	5.158[f]	0.26	0.99	2.04	3.28
			38.685[e]	7.737[g]	0.26	0.99	2.04	3.28
Co	^{60}Co	IVb	0	0	−0.01	−0.05	−0.10	−0.19
			4620.6[a]	4620.6[a]	−0.01	−0.05	−0.10	−0.19
			46206[b]	46206[b]	−0.01	−0.05	−0.10	−0.19
			231032[c]	46206[b]	−0.01	−0.05	−0.10	−0.19
			462064[d]	92413[f]	−0.01	−0.05	−0.10	−0.19
			693096[e]	138619[g]	−0.01	−0.05	−0.10	−0.19
Mo	^{99m}Tc	IId	0	0	−0.30	−0.62	−0.91	−1.15
			6.594[a]	6.594[a]	−0.04	−0.12	−0.21	−0.30
			65.94[b]	65.94[b]	0.00	0.00	0.00	0.00
			329.7[c]	65.94[b]	0.00	0.00	0.00	0.00
			659.4[d]	131.88[f]	0.00	0.00	0.00	0.00
			989.1[e]	197.82[g]	0.00	0.00	0.00	0.00

The slope $k = -0.005$ (IC-40 channel) is assumed in the calculation of the F_{sat}

[a] 0.1 × $T_{1/2}$ of the nuclide

[b] 1 × $T_{1/2}$ of the nuclide

[c] 5 × $T_{1/2}$ of the nuclide

[d] 10 × $T_{1/2}$ of the nuclide

[e] 15 × $T_{1/2}$ of the nuclide

[f] 2 × $T_{1/2}$ of the nuclide

[g] 3 × $T_{1/2}$ of the nuclide

control rod positions for different irradiations of about 20 h from the operators' logbook are shown in Table 1 for different reactor core configurations. It can be seen that movement of the control rod (labelled R in Fig. 1) is about 160 steps, where step 200 is at the top and step 900 is at the bottom of the active core.

The distribution of the reactor power during operation for the same irradiations (5 randomly chosen for core No. 176 and No. 189), shown in Table 1, is presented in Fig. 2. We took into consideration the similarity of the reactor power variation to calculate the slope of the neutron flux (signed as k in Fig. 2). The average slope $k = –0.005$ and $k = –0.004$ is calculated from 5 irradiations over the time interval from 5 to 20 h for core No. 176 and core No. 189, respectively. Figure 3 shows the reactor power electrometer reading from the safety channel for core No. 176.

During the first 3 or 4 h of operation the true power of the reactor increases slowly and then decreases almost linearly. This is due to the fact that the instrumentation focuses on the reading of one detector (i.e. the linear channel). Due to the changes of the fuel and coolant temperatures and xenon buildup the core reactivity changes, requiring the re-positioning of control rods. With the reading of the linear channel fixed, the power redistribution results in a change of the true average core power (see Fig. 4).

Calculated correction factors F_{sat} (in %) based on observed temporal (time-dependent) variation of the neutron flux in the CF of TRIGA reactor for long irradiation are presented in Table 2. Table 3 shows some examples of the reduction of F_{sat} based on different cooling and measurement times. This reduction is particularly evident for more complex activation-decay types (e.g. type IId) [4].

Conclusions

The conclusions from our study can be summarised as follows:

Previous measurements on the TRIGA reactor at the JSI before reconstruction indicate that time-dependent variations in the flux are more or less random and depend on the actions of the operator [7].

After reconstruction the upgraded instrumentation resulted in operation modes where the position of the compensating control rod is normally fixed and the automatic control system adjusts the position of the regulating rod to keep reactor power constant (as measured from the linear channel).

It is well known that due to control rod movement, temperature distribution and xenon buildup a redistribution of neutron flux in the core occurs, resulting in a discrepancy in the power measured at different locations by ionisation chambers. Similar changes also occur in different irradiation channels.

Time-dependence of the flux variations was parameterised and correction factors for these variations were calculated for a number of analytically-interesting nuclides for various irradiation channels of the CF in the TRIGA reactor at the JSI. The correction results in a reduction of the measured specific activity (current $F_{sat} < F_{sat}$ at the end of irradiation), depending on the cooling and the measurement times and also on the specific activation-decay scheme.

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