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Re-characterization of irradiation facilities for k_0-NAA at RPI after conversion to LEU fuel and re-arrangement of core configuration

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ABSTRACT

The 1 MW pool-type Portuguese research reactor (RPI) was converted from using highly enriched uranium (HEU) to using low enriched uranium (LEU) fuel in September 2007. Although the new LEU core has the same number of fuel assemblies as the first HEU core, it has three assemblies less than the last HEU core and also has a different arrangement of the beryllium reflectors. Therefore, the primary irradiation positions that are used with the k_0-based neutron activation analysis (k_0-NAA) at the RPI were redefined and characterized. The neutron spectrum parameters of the deviation of the epithermal neutron spectrum distribution from the $1/E$ shape (α), the thermal to epithermal neutron flux ratio (f), the thermal to fast neutron flux ratio (f_F) and the neutron temperature (T_n) were determined by using a combination of Au, Lu, Ni and Zr monitors. The experimental results of the neutron spectrum parameters obtained in this study are compared to the results were obtained prior to the refuel conversion in the applicable irradiation positions. The synthetic multi-element standard SMELS was used to verify the accuracy of k_0-NAA applied to practical samples using the newly measured neutron spectrum parameters.

1. Introduction

The Portuguese research reactor (RPI) is a 1 MW open-pool reactor. The core of the RPI was converted to low enriched uranium fuel (LEU) in September 2007. The reactor now uses the 19.75% LEU fuel in the form of U_3Si_2–Al with a density of 4.8 g U/cm^3. Although the new LEU core has the same number of fuel assemblies as the first HEU core, it has three assemblies less than the last HEU core and also has a different arrangement of beryllium reflectors [1]. Table 1 provides a summary of the key design features of the HEU and LEU fuel.

The RPI does not have a standard core. As it burns less than 100 g of ^{235}U per year, core management is reduced to shuffling of assemblies for a more homogeneous burn-up, with eventual re-arrangement of the Be reflectors, followed by the addition of fresh assemblies when necessary. Fig. 1 shows a schematic view of the current core configuration in the RPI reactor. The blocks marked "DA" in Fig. 1 are hollow dummy assemblies that allow irradiations inside; their function is to preserve the cooling conditions of the fuel assemblies in the border of the core.

The irradiation facilities that are used to support neutron activation analysis (NAA) at the RPI include: (1) a fixed pneumatic transfer system, with a terminal in the hot chemistry room; (2) a movable fast pneumatic transfer system (SIPRA) for short irradiations for which the sample being irradiated can be covered with Cd; (3) several core cells, with thermal neutron flux up to 3×10^{13} cm^{-2} s^{-1}. The samples from long irradiations are handled in the hot chemistry laboratory that is located in the reactor hall and is equipped with several tools for radiation protection and transportation containers.

The significant changes introduced after the core conversion triggered a new evaluation of the irradiation positions and a determination of the neutron spectrum parameters for use in the k_0-NAA method [2,3]. The methodology relies on the description of activation rate as the capture probability per atom per unit time p, which is treated as the sum of the integral of the Maxwell–Boltzmann spectrum times the cross-section, which may be non-$1/v$, plus the integral over the epithermal spectrum times the resonances and the uranium fission spectrum [4]:

$$p = \Phi_t g(T_n)\sigma_0 + \Phi_e I_0(\alpha) + \Phi_f \sigma_f$$

where Φ_t, Φ_e and Φ_f are the thermal, epithermal and fast neutron fluxes, respectively; $g(T_n)$ is the Westcott factor depending on the neutron temperature used to correct for the non-$1/v$ nuclides according to the modified Westcott formalism [4,5]; $I_0(\alpha)$ is the resonance integral corrected with the deviation of epithermal neutron spectrum from $1/E$ shape; and σ_f is the ^{235}U fission neutron cross-section. However, in the k_0-NAA application, the ratios of the thermal to epithermal neutron flux (f) and the thermal to fast neutron flux (f_F) used for the correction of interference reactions induced by fast neutrons are replaced for

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the individual neutron fluxes. Therefore, the primary neutron spectrum parameters that are considered for corrections in k_0-NAA include α, f, f_F and T_n. The neutron spectrum parameters are determined by irradiating and measuring a set of monitors such as Au, Lu, Ni and Zr in metallic or alloyed foil and/or wire. The nuclear data of Au and Zr monitors with the effective resonance neutron energy $\overline{E}_r$ varying over a large interval (from 5.65 eV for ^{197}Au to 6260 eV for ^{94}Zr) allow checking the consistency of α over the whole epithermal neutron region at the irradiation channel.

The synthetic multi-element standard SMELS [6] was used to verify the accuracy of k_0-NAA applied to practical samples using the newly measured neutron spectrum parameters in which the k_0-IAEA program [7] was applied for the calculation of the mass fraction of element. In order to evaluate the laboratory performance, the u-score test [8] was used in this study. The u-score is calculated according to the following equation: u-score$=(x_{lab}-x_{ref})/\sqrt{u_{lab}^2+u_{ref}^2}$, where x_{lab}, u_{lab}, x_{ref} and u_{ref} are the laboratory results, overall/combined standard uncertainties, the assigned values and standard uncertainties, respectively. The results of the laboratory are interpreted according to the 5 possible evaluation classes as follows: (1) $u<1.64$, the laboratory result does not differ significantly from the assigned value; (2) $1.96>u>1.64$, the laboratory result probably does not differ significantly from the assigned value; (3) $2.58>u>1.96$, it is not clear whether the laboratory result differs significantly from the assigned value; (4) $3.29>u>2.58$, the laboratory result is probably significantly different from the assigned value; (5) $u>3.29$, the laboratory result is significantly different from the assigned value.

Table 1
Summary of HEU and LEU key design data used for the RPI reactor (shortened for this study concern).

Design data	HEU	LEU
Fuel meat composition	U–Al alloy	U_3Si_2-Al
Uranium enrichment (nominal) (%)	93.2	19.75
Uranium density in fuel meat (g/cm^3)	0.83	4.8
Number of standard assemblies (Initial Core)	7	7
Mass of ^{235}U per standard assembly (g)	265	376
Number of plates per standard assembly	18	18
Number of control assemblies (Initial Core)	5	5
Mass of ^{235}U per control assembly (g)	147	209
Number of plates per control assembly	10	10

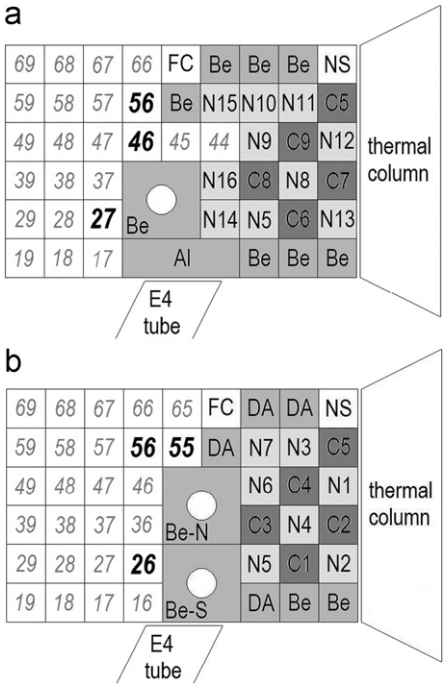

Fig. 1. Core configuration of the RPI reactor (a) before (HEU) and (b) after (LEU) fuel conversion. The cells with bold numbers were investigated in this study, i.e. Cells 27, 46 and 56 for HEU and Cells 26, 55 and 56 for LEU core configuration. (N: standard fuel assemblies, C: fuel assemblies with control or regulating rods, Al: aluminum, NS: neutron source, FC: fission chamber, Be, BeN and BeS: beryllium reflectors, DA: dummy assembly.)

2. Experimental

Three sets of monitors were put into high purity polyethylene vials and loaded into containers for each irradiation channel. The channels are here designated SIPRA (corresponding to Cell 27 in the last HEU core and Cell 26 in the new LEU core—see Fig. 1), the positions that are normally used for the irradiation of biological samples (Cell 45 for HEU core and Cell 55 in the LEU core—see Fig. 1) and the positions that are normally used for the irradiation of air filters (Cell 56 in both cores). Although the cells in the LEU and HEU cores are not necessarily the same, they have adequate characteristics for NAA purposes.

Table 2 shows the irradiation, decay and counting times for the monitors. Typically monitors with masses of 15 mg for Al–0.1% Au foil (IRMM-530R) and Al–0.1% Lu (wire), 20 mg for pure Ni (wire) and Zr (foil) were irradiated for 15 min and allowed to decay for 3–5 h for ^{65}Ni and ^{176m}Lu, 1 day for ^{97}Zr and 3 days for ^{198}Au, ^{177}Lu, ^{95}Zr and ^{58}Co prior to counting on the gamma-ray spectrometers.

Table 2
The irradiation, decay and counting times for the monitors with Al–0.1% Au and Zr in foil shape and Ni and Al–0.1% Lu in wire geometry (combination: monitors put together during the measurement).

Irradiation time (monitor, mass)	Decay time	Counting time (combination)	Measured radionuclides ($T_{1/2}$, γ-rays in keV)
15 min (Au and Lu, ~15 mg)	~3–5 h	0.5–1 h (1 h)	^{65}Ni (2.5 h, 366.3, 1115.5, 1481.8) ^{176m}Lu (3.6 h, 88.4)
	~1 d	1–2 h (2 h)	^{97m}Nb (60 s, 743.4)* ^{97}Nb (16.7 h, 657.9)
(Zr and Ni, ~20 mg)	~3 d	0.5–3 h (5 h)	^{198}Au (2.7 d, 411.8) ^{177}Lu (6.7 d, 112.9, 208.4) ^{95}Zr (64 d, 756.7) ^{95}Nb (64 d, 765.8) ^{58}Co (70.8 d, 810.8)

* Nuclide ^{97m}Nb is decayed from nuclide ^{97}Zr with half-life of 16.7 h.

Table 3
The results of the determination of neutron spectrum parameters at the irradiation facilities of the RPI reactor (Φ_{th}: thermal neutron flux in $cm^{-2}s^{-1}$).

Parameters	Irradiation position for SIPRA		Irradiation position for aerosols		Irradiation position for biological samples	
	HEU (Cell 27)	LEU (Cell 26)	HEU (Cell 46)	LEU (Cell 55)	HEU (Cell 56)	LEU (Cell 56)
Φ_{th} ($\times 10^{12}$)	2.31 ± 0.25	2.68 ± 0.25	7.39 ± 0.25	8.41 ± 0.09	2.79 ± 0.25	2.36 ± 0.15
$R_{Cd,Au}$	–	8.7	–	5.9	–	8.2
α	0.050 ± 0.018	0.025 ± 0.015	0.050 ± 0.020	-0.040 ± 0.016	0.030 ± 0.017	0.040 ± 0.018
f	127 ± 10	113 ± 12	78 ± 8	82 ± 9	98 ± 10	105 ± 11
f_F	24.84	21.53	16.63	15.29	13.90	12.72
T_n (K)	293 ± 10 (19.9 °C)	297 ± 10 (23.9 °C)	329 ± 15 (55.9 °C)	330 ± 16 (56.8 °C)	(44 °C)*	318 ± 10 (44.8 °C)

* The neutron temperature was determined in HEU configuration by Mustra et al. [12]; Uncertainty is presented as the standard deviation of the results.

The monitors were measured at a distance of at least 10 cm from the face of calibrated HPGe detectors [9] with the FWHM approximately 1.85 keV at 1332.5 keV and relative efficiencies of 20–30%. The counting time for each monitor ranged from 0.5 to 3 h in order to obtain a minimum of 10,000 counts in the peak of interest. After counting each monitor, a selected combination of monitors was re-measured as indicated in Table 2 with counting times ranging from 1 to 5 h. As for evaluation of the neutron spectrum parameters determined in this study, the SMELS consists of three types of materials: Types I, II and III with elements forming short, medium and long-lived radionuclides, respectively, which were weighed around 100–150 mg and put into pure polyethylene vials in preparation for irradiation. SMELS Type I was irradiated for 100 s on SIPRA at a thermal neutron flux of about $2.7 \times 10^{12}\,cm^{-2}\,s^{-1}$ and allowed to decay for 10 s prior to counting of 200 s on an HPGe-coupled detector. After about several hours the samples were re-measured in order to acquire medium-lived radionuclides. The SMELS Types II and III were irradiated in Cell 55 for 1 h and Cell 56 for 5 h (thermal neutron fluxes of about 8.4×10^{12} and $2.4 \times 10^{12}\,cm^{-2}\,s^{-1}$, respectively) and allowed to decay for 2–3 days and 3 weeks prior to the first and second measurements using ORTEC HPGe detector-based automatic sample changers. It should be noted that Au was added/spiked in all three SMELS types and Zr in type III. Therefore, type III should also be used as an internal standard to check α and f values.

The k_0-IAEA program was used for the calculation of neutron spectrum parameters. However, for comparative purposes, the calculation of α and f factors was also performed by an iterative linear regression approach using an MS-Excel spreadsheet [10] and the determination of f_F and T_n factors was described in Ref. [11]. The gamma-ray spectrum analysis was carried out by the k_0-IAEA program for use with MS-Excel spreadsheet method as well.

3. Results and discussion

The results were obtained using two separate calculation approaches for α and f factors: an iterative linear regression using an MS-Excel spreadsheet and a mathematical numerical analysis using the k_0-IAEA program. Both methods were in rather good agreement with each other. The α values obtained by the spreadsheet method were a little lower than those values obtained by the k_0-IAEA program, with differences of 3.4% at SIPRA and 2.9% at Cell 55. The f values obtained by the "bare bi-isotopic monitor" method with the reactions $^{94}Zr(n,\gamma)^{95}Zr$ and $^{96}Zr(n,\gamma)^{97}Zr/^{97m}Nb$ indicated larger values than those obtained by the k_0-IAEA program, with differences of 7.6% at SIPRA and 9.1% at Cell 55. It should be noted that α and f factors were also calculated by k_0-IAEA program using SMELS Type III with results higher by about 26.3% for α and 11.7% for f than those results using traditional monitors.

The results of the determination of neutron spectrum parameters at the irradiation positions of the RPI are displayed in Table 3. The data in Table 3 show that although the irradiation positions at SIPRA and the position for irradiation of aerosol (Cell 46 before and Cell 55 after conversion) were not the same, they are comparable. It is interesting to note the variations in the other parameters, mostly as a result of the different arrangement of fuel and reflectors. The values of α and f in Table 3 indicate that the most remarkable fact is that the α decreased by 50.0% at SIPRA, increased by 33.3% at Cell 56 and changed into a negative value at position for irradiation of aerosol, after conversion to LEU fuel. The f value decreased by 11.0% at SIPRA and slightly increased within 5–7% at Cells 55 and 56. The changes of α and f must be taken into account when applying k_0-NAA, particularly in determining elements forming radionuclides with Q_0-values much different from ^{198}Au monitor ($Q_0 = 15.7$). This can be illustrated most clearly with the short-lived radionuclides such as ^{49}Ca, ^{51}Ti and ^{52}V having Q_0-values of 0.45, 0.67 and 0.55, respectively.

No significant changes are observed in the temperature at the irradiation positions in the new LEU core in the studied irradiation channels, as the measured differences are less than 2%. The LEU fuel configuration also results in different ratios of thermal to fast neutrons (f_F) in the irradiation sites. The ratios were measured to be 21.53, 15.29 and 12.72 with the LEU configuration and 24.84, 16.63 and 13.90 with the HEU configuration in SIPRA (Cell 27/26), Cell 46/55 and Cell 56, respectively. The ratio f_F decreases, which implies the fast neutron flux increases. This effect creates increasing interfering reactions: the so-called threshold reactions such as (n, p), (n, n'), etc. in the k_0-NAA method. Therefore, the f_F factor must be used when applying k_0-NAA to the practical samples.

Table 4 shows the mean result of six determinations in terms of mass fractions of element as compared to the assigned values in SMELS Types I, II and III. The bias of the results—using the neutron spectrum parameters obtained in this study—in comparison with the assigned values was less than 5% with the u-score values less than |1.64| for most elements except for Sr in Type III, which was equal to 1.64. It should be noted that the use of Au in SMELS as an internal standard for flux monitor by k_0-IAEA program was not successful since the software does not accept two actions happened simultaneously, i.e. the same spectrum for comparator and the sample. The problem should be overcome by improving the experiment with more measurements of SMELS sample so the software regards the individual spectra of the sample as separated sample spectra.

4. Conclusions

The re-characterization of the neutron spectrum parameters (thermal neutron flux, α, f, f_F and T_n) was performed for the RPI reactor after the conversion to LEU fuel and the re-arrangement of

Table 4
Elemental concentrations (mg/kg) in SMELS Types I, II and III determined by k_0-NAA established at RPI/ITN using k_0-IAEA software.

Element	SMELS Type I				SMELS Type II				SMELS Type III			
	$x_{ref} \pm u_{ref}$	$x_{lab} \pm u_{lab}$ (n=6)	x_{lab}/x_{ref} Ratio	u-Score	$x_{ref} \pm u_{ref}$	$x_{lab} \pm u_{lab}$ (n=6)	x_{lab}/x_{ref} Ratio	u-Score	$x_{ref} \pm u_{ref}$	$x_{lab} \pm u_{lab}$ (n=6)	x_{lab}/x_{ref} Ratio	u-Score
As	–				92.3 ± 1.8	91.1 ± 3.7	0.99	−0.30	–			
Au	82.7 ± 0.9	80.9 ± 1.8	0.98	−0.90	3.93 ± 0.04	3.81 ± 0.08	0.97	−1.39	0.901 ± 0.008	0.921 ± 0.018	1.02	1.04
Br	–				157 ± 3	154 ± 6	0.98	−0.52	–			
Ce	–				15600 ± 400	15918 ± 812	1.02	0.35	–			
Cl	4330 ± 85	4505 ± 161	1.04	0.96	–				–			
Co	–				–				24.3 ± 0.2	23.7 ± 0.4	0.97	−1.49
Cr	–				–				86.7 ± 1.3	89.4 ± 2.4	1.03	0.98
Cs	897 ± 19	856 ± 43	0.95	−0.87	–				20.80 ± 0.17	21.8 ± 0.7	1.05	1.39
Cu	3930 ± 60	3917 ± 119	1.00	−0.10	–				–			
Fe	–				–				8200 ± 95	7885 ± 195	0.96	−1.45
I	152 ± 3	155 ± 6	1.02	0.45	–				–			
In	–				–				462 ± 10	486 ± 21	1.05	1.05
La	265 ± 5	255 ± 11	0.96	−0.83	–				–			
Mn	113.9 ± 1.7	112.4 ± 3.6	0.99	−0.38	–				–			
Mo	–				5170 ± 125	5442 ± 290	1.05	0.86	–			
Pr	–				1193 ± 19	1147 ± 35	0.96	−1.15	–			
Sb	–				172 ± 4	169 ± 7	0.98	−0.43	51.2 ± 0.7	49.7 ± 1.4	0.97	−0.95
Sc	–				–				1.140 ± 0.016	1.162 ± 0.035	1.02	0.58
Se	–				–				131 ± 3	137 ± 5	1.05	1.03
Sr	–				–				8150 ± 100	8490 ± 182	1.04	1.64
Th	–				3670 ± 90	3851 ± 189	1.05	0.87	26.2 ± 0.5	25.7 ± 0.9	0.98	−0.50
Tm	–				–				23.3 ± 0.4	24.0 ± 0.7	1.03	0.89
V	39.0 ± 0.8	38.0 ± 1.7	0.97	−0.53	–				–			
Yb	–				187 ± 5	180 ± 9	0.96	−0.70	20.7 ± 0.3	21.3 ± 0.4	1.03	1.28
Zn	–				6570 ± 100	6916 ± 211	1.05	1.48	618 ± 6	613 ± 16	0.99	−0.29
Zr	–				–				4580 ± 50	4490 ± 99	0.98	−0.81

x_{lab} and u_{lab}—the laboratory results and overall/combined standard uncertainties, respectively; x_{ref} and u_{ref}—the assigned values and standard uncertainties, respectively; n—replicate number.

core configuration at the irradiation positions that are used for the k_0-NAA method. The thermal neutron fluxes, together with other neutron spectrum parameters, are suitable for short irradiations at SIPRA and long irradiations for aerosol and biological samples at Cell 55 and Cell 56, respectively. The f values changed by 11.0%, 5% and 7% at SIPRA and Cells 55 and 56, respectively, and the fast neutron ratios (f_F) decreased by 13.3%, 8.1% and 8.5% compared to the HEU configuration. Finally, the analysis of SMELS reference material demonstrated that the neutron spectrum parameters obtained by this study are applicable for practical analyses using the k_0-NAA method at the RPI after conversion to LEU fuel and re-arrangement of core configuration.

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