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RADIONUCLIDE ANALYSIS OF THE IEA-R1 POOL WATER

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ABSTRACT

IEA-R1 is a 5MW pool-type research reactor built in the late 1950's. In the last years, it operates at 4.5MW for 8h on Mondays, Tuesdays and Wednesdays. In every day of operation, a sample is taken from the pool water both before starting the reactor and at the end of the day and analyzed in an HPGe detector, in order to verify for possible problems with the fuel elements or other issues. In this work, the results obtained in these analyses spanning for some months are discussed regarding the radionuclides frequently identified and the dependence of their activities with time.

1. INTRODUCTION

IEA-R1 is the oldest nuclear reactor in operation in Brazil, and also the largest research reactor in the country. It is an open pool type reactor built in the late 1950's with a nominal power of 5 MW; in the last years, though, it operates 8.5 hours a day (from 7h30 to 16h, mostly), 3 days a week (from monday to wednesday), at 4.5 MW.

The nuclear reactor pool water is an essential element in the safe operation of the reactor [1] and, as it comes into close contact with the reactor core, where the neutron flux is highest, it must be highly demineralized in order to reduce the formation of radionuclides due to neutron irradiation. Moreover, the pool water is an excellent indicator of problems in the fuel elements, as any leakage will definitely introduce fission products into the pool water.

As part of the IEA-R1 quality assurance process, its pool water is checked daily for the presence of radionuclides that may indicate problems either in the fuel elements or in the water purification process. For this purpose, twice in each day of operation, 100 mL of pool water are collected, once at 7 A.M., before the reactor is turned, on and again at 3 P.M., close to the end of the daily operation. This water is then gamma-counted in an HPGe detector, and the spectrum is checked for the presence of extraneous gamma peaks. The water collection is performed using a small pump connected to a faucet, and the water is sampled directly above the core, at 1 m depth (so, at approximately 5 m from the core itself) into a plastic vessel.

In this work, the pool water samples were counted in a characterized HPGe detector, in order to identify and quantify the most relevant radionuclides found; the measurements span for two months, to allow for a discussion on dependence of the activities with time.

2. EXPERIMENTAL PROCEDURE

After the water samples were collected by the reactor staff and analyzed in the regular way (i.e., in a conventional HPGe detector), as required by the reactor's quality control, the same samples were then counted a second time, in a characterized HPGe detector, in order to obtain precise values for the activities of the radionuclides present in the sample; due to this process, the water samples were always counted approximately 35-40 minutes after sampling.

The detector used was a 40% Canberra Extended Range HPGe with a carbon composite window; the detector was characterized at the factory, so its efficiency for extense samples can be accurately calculated by means of the LabSOCS software [2].

Samples were counted for 1500 s (Live Time) and the spectra were analyzed using Canberra's Genie-2000 software. The first step of nuclide identification was performed using Genie-2000's *NID* (Nuclide IDentification) routine with a specifically-tailored nuclide library that includes most common uranium fission and activation products, plus a series of neutron activation products that could be present in the water. As the software failed to identify some nuclides that were clearly present, a second step was then performed by manually locating the most intense unassigned gamma transitions and assigning them to the corresponding radionuclides. The detection efficiency used for each peak was the one obtained in the LabSOCS software, and the gamma intensities were obtained from [3]. Using these data, the activity for each radionuclide was determined by eq. 1, where A is the total activity in the 100 mL sample, *cps* is the counts per second for a specific transition, $\varepsilon(E)$ is the efficiency for that energy and I_γ is the transition intensity. When more than one transition was assigned to the same radionuclide, a σ^{-2}-weighted average was used. It is important to stress, too, that no decay correction was applied.

$$A = \frac{cps}{\varepsilon(E) \cdot I_\gamma}. \tag{1}$$

3. RESULTS AND DISCUSSION

The main nuclides identified in the water samples are shown in Table 1, together with their half-lives and the gamma transitions used in their identification and quantification. It must be stressed that, as the analysis took place approximately 35-40 minutes after the water was collected from the pool, short-lived radionuclides known to be present in reactor pool water as ^{16}N and ^{28}Al, for instance [1], could not be observed. Moreover, the 511 keV peak from the electron-positron annihilation was observed but can't be positively assigned to any particular radionuclide as it may arise from any β^+ decaying nuclide and, to a lower extent, also from high energy gamma transitions that may produce positrons via the pair production effect.

Table 1: Essential data about the radionuclides identified - nuclear data was retrieved from [3].

Radionuclide	Transitions Used (keV)	Half-Life
^{24}Na	1368.6[1]	14.96 h
^{27}Mg	843.7, 1014.4	9.46 min
^{38}Cl	1642.7, 2167.4	37.2 min
^{41}Ar	1293.6	109.3 min
^{54}Mn	834.8	312.3 days
^{56}Mn	846.8, 1810.8, 2113.1	2.58 h
^{58}Co	810.8	70.86 days
^{60}Co	1173.2, 1332.5	5.27 years
^{65}Zn	1115.5	244.3 days
^{110m}Ag	657.8, 706.7, 763.9, 884.7, 937.5, 1384.3, 1505.0	249.8 days
annihilation	511	*undefined*[2]

[1] The highly intense 2754.0 keV transition was above the detector's operating range ans couldn't be found; the single- and double-escape peaks related to it were observed, but not used in the quantification.
[2] The 511 keV transition is a consequence of any electron-positron annihilation process so it cannot be positively associated with any individual nuclide - it can, though, be a good indication of β^+ decaying nuclides.

Figure 1 shows the activity variation of the nuclides identified; the left figure shows the results obtained with the morning samples, while the right side shows the results for the afternoon ones.

As a general trend, as the reactor stays off for more than 15 hours every night, the pre-start (morning) samples present mostly longer-lived isotopes, while some shorter-lived ones can be found in the afternoon samples; also, as the reactor operates from monday to wednesday and remains off from wednesday afternoon to monday morning, a weekly variation can be seen – it is most noticeable in the ^{24}Na activities obtained in the morning samples. It can also be noted that the activity of the longer-lived isotopes tend to grow with time – this can be clearly observed in the ^{58}Co and ^{60}Co activities.

Looking at the radioisotopes identified, some works in the literature [4, 5, 6] indicate that ^{24}Na and ^{27}Mg are known to be produced in reactors by means of the $^{27}Al(n,\alpha)^{24}Na$ and $^{27}Al(n,p)^{27}Mg$ nuclear reactions, as Al is the main component of the fuel rod casing – the directly-produced ^{28}Al has a short (2.2 min) half-life, thus would not be seen in the present measurements. ^{38}Cl and ^{41}Ar are also known to be common in reactor pool water, arising from the neutron activation of Cl and Ar, respectively. The manganese, cobalt and zinc isotopes come from the activation of steel components (used in the pool

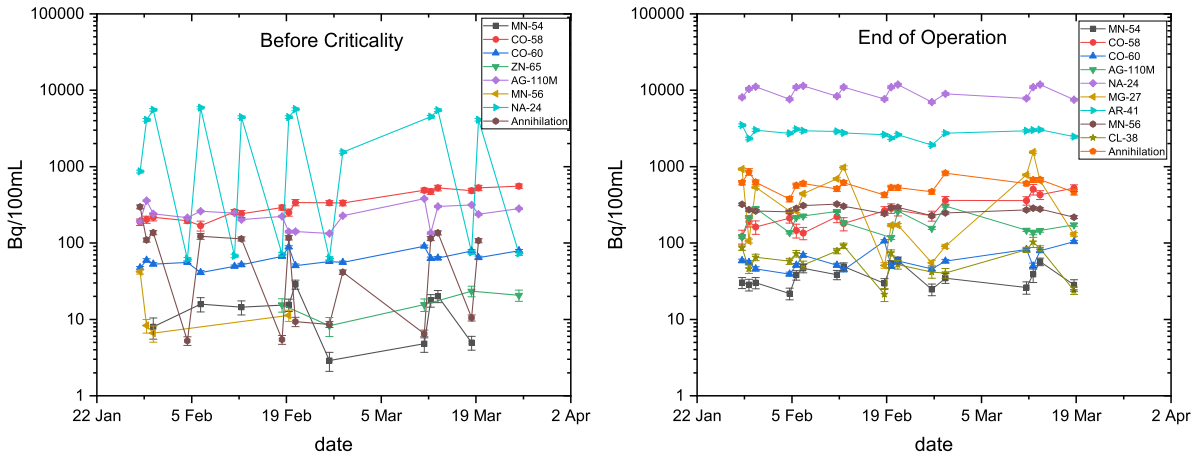

Figure 1: Variation of the nuclide activity with the collection date; on the left are the results related to the samples collected just before reactor start, and on the right the samples collected with the reactor in operation, close to the shutdown time.

inner surface and in the primary cooling tubing) and ^{110m}Ag comes from the activation of silver, used in the reactor's control rods.

Some radionuclides with very low activity were identified in a small number of the samples, but were not included in the present analysis and would deserve a closer inspection, possibly with a much longer counting time. Among them are ^{59}Fe, from the activation of the steel, and also some In isotopes, from the control rods.

A very important result is that no isotope directly related to the uranium irradiation (^{239}Pu, ^{137}Cs, among others) was found, indicating that the integrity of the fuel rods is preserved.

4. CONCLUSIONS

In this work the IEA-R1 reactor water was analyzed, and the most active radionuclides present were identified. These nuclides were found to be arising from the activation of atmospheric elements, as well as from neutron irradiation of the reactor's components – mainly from the aluminum of the fuel rods, the steel from the pool surface and primary cooling tubing, and silver from the control rods. On the other hand, no nuclides associated with uranium activation of fission were found, indicating that the fuel element integrity is preserved.

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