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Accounting for Radionuclides Generated in Reactor IEA-R1

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1. Introduction

The IEA-R1 reactor of the Institute for Energy and Nuclear Research (IPEN), installed at the Research Reactor Center (CERPq) is an open pool type reactor. Before going to the pool, the water is previously demineralized to reduce the formation of radionuclides by irradiation with neutrons, however, circulation through the system can lead to events that cause the dragging of materials, both from the air and from the fission products, which can form fission products in the pool water during operation. As part of the quality assurance process for the IEA-R1 reactor, the pool water is analyzed twice a day to check for the presence of radionuclides that could indicate problems, whether in the fuel elements or in the water purification process.

In the case of the IEA-R1 reactor, the pool water is purified by treatment using a system of carbon filters and ion exchange resins. Every 750 hours of operation of the reactor with a power of 4.5 MW, it is necessary to carry out the resin regeneration procedure, in which approximately 3500 liters of water are used in the process that involves washing the coal, recovering the resins, washing the resin and final washing of the treatment and retreatment system with a solution of sulfuric acid and 4% sodium hydroxide. The water used in this process is discarded and stored in a containment tank.

The IEA-R1 primary cooling system consists of a pool, piping, decay tank, pumps, heat exchangers, distributor, valves and frames. The reactor core is cooled by downflow. Heat removal from the reactor is based on primary and secondary cooling systems. Water from the primary system pool is pumped downward through the fuel elements to remove fission heat from the core. Water then flows through the decay tank to slow activity before entering the heat exchanger, which transfers heat to the secondary cooling system. The secondary system circulates the heated water to the cooling tower which dissipates the heat to the atmosphere. Water from the primary circuit returns to the reactor pool through diffusers in the lower region of the pool. During the cooling process, there is a deliberate loss of water, therefore, this stage also generates radioactive waste that is stored in the same containment tank as the waste generated during resin regeneration.

The containment tank used to store the radioactive waste mentioned above has a capacity of 4 m3, which, when full, is taken to a second containment tank.

A work described by ZAHN et al. [1] identified the presence of nuclides that could be associated with the activation of atmospheric elements, as well as with the neutron activation of reactor components − mainly the aluminum of the fuel elements, the steel of the internal surface of the pool and the primary circuit piping, and the silver of the control rods. However, no nuclides associated with uranium activation or fission were found, indicating the preservation of the integrity of the fuel elements.

However, work carried out between 2003 and 2019 [2] aimed at detecting and identifying radionuclides in the reactor pool, indicated that the radionuclides Ag-110m, Co-58, Co-60, Cr-51, I -131, Mn-54, Na-24, Np-239, Te-132, W-187 and Zn-65 are usually detected, while Cs-137, Ba-140/La-140 and Ru-103 are rarely detected. This work aims to analyze the water from the IEA-R1 reactor on operating days and the waste from the two containment tanks to verify which radionuclides were found, the radioactivity and their possible origin.

2. Methodology

Pool water samples (APR) were collected on reactor operating days, totaling 12 samples, collected twice a week.

The waste was collected directly from the containment tanks (T1 and T2) with the help of scoops and polyethylene pots previously sanitized with 1% nitric acid. Because they were submerged in water, after collection, the waste was filtered in AP20 filter in glass microfiber and both water and solids were analyzed separately. Two water and waste samples were collected from tank one (T1H2O and T1REJ) and two (T2H2O and T2REJ).

Water samples were prepared for measurement by manual homogenization and transfer of a volume of 250 ml to a polyethylene bottle with a rectangular section, which is a standardized geometry in the laboratory. The same was done in solid residue with approximately 1 g. The measurement system consists of an Intertechnique high purity germanium (HPGe) detector with a horizontal configuration.

3. Results and Discussion

The uranium fission products Co-60, Np-239, Ru-103 and Ru-106 were found in the APR samples, but of these, Co-60 and Ru-106 appeared both in the tank water and in their respective waste, as well as the radionuclides Cs-137 and Zr-95. Uranium fission products found in the sample may indicate that the integrity of the fuel elements may not be preserved. The activation products Am-241, Ce-144, Cr-51, Cs-136, Ni-59 and Zn-95 appeared only in the APR samples, while Ag-110m, Co-58, I-129 and Mn-54 appeared both in the APR samples and in the water and waste from the two containment tanks. The Ag-108m radionuclide was detected only in the solid waste from tank 2. Table I show maximum and minimum values found in samples analyzed.

The activation products found may be associated with the neutron activation of reactor components − mainly the aluminum in the fuel elements, the steel on the internal surface of the pool and the primary circuit piping, and the silver in the control bars.

It is noted that in solid waste from tanks, the measured radioactivity of radionuclides is greater in the order of 10 to 1000 times in relation to water from the respective waste.

Of the radionuclides mentioned, these were also found in the reactor pool water in a study described by Máduar et al. [2], in the order of 10^2 to 10^3 Bq/L.

Table I: Minimum and maximum values of radioactivity of radioniclides, in Bq/g, found in the water of the reactor pool (APR), waters and streams of tank 1 (T1H2O and T1REJ) and tank 2 (T2H2O and T2REJ)

	Ag-108m	**Ag-110m**	**Am-241**	**Ce-144**	**Co-57**	**Co-58**	**Co-60**
APR	-	1.3E-02 - 8.9E-02	3.5E-3 - 8.2E-3	<6.910E-03 - 2.7E-01	-	4.2E-02 - 8.7E-02	1.2E-01 - 2.4E-02
T1H2O	-	-	-	-	-	-	6.43E-01 - 1.8E+00
T2H2O	-	<1.076E-03 - 4.28E-03	-	-	-	-	4.06E-02 - 5.85E-01
T1REJ	-	<26E-01 - 500E-01	-	-	-	-	2.45E+03 - 7.79E+04
T2REJ	49E-01 - 220E-01	1E+00 - 30E+00	-	-	1.5E-01 - 05.7E-01	1.5E-01 - 5.-014	3.17E+03 - 7.76E+03
	Cr-51	**Cs-136**	**Cs-137**	**I-129**	**Mn-54**	**Ni-59**	**Np-239**
APR	<4.17E-02 - 1.9E-01	<7.41E-0.3 - 3.5E-03	-	-	6.8E-03 - 1.1E-02	7.93E-03 - 1.3E-01	<4.46E-03 - 7.7E-03
T1H2O	-	-	2.91E-02 - 1.74E-02	-	2.07E-02 - 1.03E-02	-	-
T2H2O	-	-	3.25E-02 - 3.00E-02	-	<7.17E-04 - 8.53E-03	-	-
T1REJ	-	-	4E+00 - 69E+00	-	4.9E+01 - 31.3+01	-	-
T2REJ	-	-	2.1E+01 - 3.0E+01	18E-01 - 20E-01	54E-01 - 149E-01	-	-
	Ru-103	**Ru-106**	**Sb-124**	**Sb-125**	**Zn-65**	**Zr-95**	
APR	<5.60E-03 - 1.0E-03	9.6E-01 - 5.00E+00	-	-	3.97E-02 - 2.8E-02	-	
T1H2O	-	2.35E-03 - 2.11E-02	-	-	-	-	
T2H2O	-	<4.75E-03 - 4.07E-03	-	-	-	-	
T1REJ	-	<13.6 - 736	-	-	-	59.8E-01 - 5.8E-01	
T2REJ	-	1.8E-01 - 7.7E-01	-	<5.5E-01 - 32E-01	-	-	

4. Conclusions

It was possible to conclude from this work that uranium fission products are formed during the operation of the reactor, as well as activation products are also formed, probably coming from the aluminum of the fuel elements, the steel of the internal surface of the pool and the piping of the primary circuit, and silver from control bars, but with low radioactivity. The liquid waste in the containment tanks had lower radioactivity than the solids, indicating that there is entrainment of particulate matter in the pipes. For future work, analyzes are suggested to determine whether there are alpha and beta (U-238, U-235, U-234, Np-237, Pu-238, Pu-239+240, Pu-242, and Th-230) emitting radionuclides in the reactor pool water in liquid and solid waste.

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