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Water quality management program for IPR-R1 TRIGA[®] research reactor

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ABSTRACT

The IPR-R1 Triga nuclear research reactor of the Nuclear Technology Development Center (CDTN) is one of the oldest reactors in operation in the world. It is a compact and inherently safe reactor that operates at a continuous power level of 100 kW with a solid homogeneous General Atomic (GA) fuel element of zirconium hydride moderator homogeneously combined with 20% enriched uranium. The reactor core is at the bottom of a tank under approximately 6.0 meters of shielding water. The long operation time of the reactor, 63 years, and the contact of the water with the core, can induce corrosive processes in the IPR-R1 reactor and affect its safe operation. To keep the water quality according to the chemical-physical recommended standards, a quality management program, as recommended by the International Atomic Energy Agency (IAEA), was implemented. The water quality management program is a guideline of good practices applied to nuclear reactors, targeting to keep their water coolants at specified physical-chemical standard. The main aim of the present work is to introduce IPR-R1 Triga's water chemistry program results from the second half of 2022 till the first half of 2023, when the installation returned to its regular activities after the Covid-19 pandemic. The physical-chemical parameters evaluated (e.g.: pH, alpha and beta radiation, electrical conductivity and gamma emitters) shows that the IPR-R1 reactor operates within recommended safety standards.

Keywords: IPR-R1 Triga Research Reactor, Water quality management program, Physical-chemical parameters.



1. INTRODUCTION

IPR-R1 Triga nuclear research reactor of the CDTN (Nuclear Technology Development Center) is one of the oldest Triga's reactor still in operation. The water has a function of biological shielding, neutron moderator and coolant [1]. The contact of the water with the fuel elements in the core can induce corrosive processes and affect the its structural integrity and he safe operation of the [2,3].

In this context, it is mandatory to maintain the chemical-physical characteristics of water coolant within recommended standards, enabling a safe and efficient operation of the nuclear installation [4]. As Triga research reactor operates for more than six decades, and as its natural aging progresses, the monitoring and controlling of the physical-chemical parameters of its water coolant becomes necessary to keep operations at safety levels; thus, minimizing corrosion mechanisms in addition to maintaining the integrity of the installation. Such goal can be achieved through a water quality management program, as described in International Atomic Energy Agency (IAEA) publication [4].

A water quality management program is a guideline of good practices applied to nuclear reactors, targeting to keep their water coolants at specified physical-chemical standards with the goal to reduce the occurrence of corrosive processes in their components, mainly fuel elements [4]. Still, according to (IAEA), "an essential part of a water quality management system is a sampling program in which the sampling frequency, collection positions and sampling procedures are well defined". Even though it is importance to nuclear installations, IAEA emphasizes that only a few publications address the rationale of water quality management in research reactors. Additionally, the IAEA points out that such approach can support research reactor operators in implementing water quality management programs in their facilities. The sharing of knowledge covering physical-chemical control programs of the cooling water of nuclear research reactors contributes for improving the operation of this kind of system [4].

In Brazil, Carvalheira *et al.* [5] presented the results of the water quality control for the Argonauta research reactor located at the Institute of Nuclear Energy (IEN) in Rio de Janeiro. The Argonauta reactor operates within the predefined limits of electrical conductivity; below 1 μ S·cm⁻¹

and pH between 5.0 and 6.0. Radiological tests did not indicate rupture in fuel elements. Still, Ticiane *et al.* [6] presented a radionuclide study about the pool water of IEA-R1 reactor located at Ipen (Institute for Energy and Nuclear Research) in São Paulo. Through this study, the authors determined the radionuclides present in IEA-R1 water, in addition to quantifying their respective radioactivity. The water was collected in two different moments: before the criticality and at the end of operation. Among the detect radionuclides are Co-58, Co-60 and Zn-65, with the respective radioactivity level (Bq·L⁻¹).

About the water quality program for the IPR-R1 Triga reactor, its first version was proposed by Auler *et al.* [7]. This program was started in 2012, and it lasted until 2015, when a new version of it was implemented. Based on the first version of the program, between 2012 and 2015, annual reports [8-10] were prepared, indicating that the physical-chemical parameters of Triga's cooling water are within the recommended limits suggested by the IAEA, and those proposed by Howell [11]. The latter was adopted by CDTN technical team as an additional reference to IAEA recommendations [4].

In 2015, Rodrigues [12] proposed the first update for such water quality management program. This update was made to approximate as much as possible the original CDTN program to the IAEA recommendations, especially regarding the monitoring frequencies for the physical-chemical parameters evaluated. More detailed explanations on the motivations for updating the original CDTN program are found in the reference [13]. Based on this update, between 2016 and 2019, annual CDTN reports [14-17] were prepared, indicating that the physical-chemical parameters of the Triga's cooling water are within the limits recommended by the IAEA and those defined by Howell [11]. Besides that, a new update for the water quality management program of IPR-R1 reactor is under discussion by CDTN technical team.

Bearing in mind that the chemical control of the cooling water of research reactors is directly related to the safety of nuclear installations, the main aim of the present work is to introduce IPR-R1 Triga's water chemistry program results from the second half of 2022 till the first half of 2023, when the installation returned to its regular activities after the Covid-19 pandemic.

2. METHODOLOGY

2.1. IAEA recommendations for the cooling water of nuclear research reactors

Table 1 shows the monitoring frequency and the recommended limits for the physical-chemical parameters of the primary cooling water of nuclear research reactors. Such recommendations are used as a reference to evaluate if the reactor operates within the recommended safety limits.

Physical-chemical pa- rameter	Recommended val- ues (limits)	Monitoring frequency	
Electrical conductivity	$< 1.0 \ \mu S \cdot cm^{-1}$	On-line; twice per week laboratory sample	
pН	between 5 and 6.5	On-line; twice per week laboratory sample	
Chlorine (Cl ⁻)	$< 0.05 \text{ mg} \cdot \text{L}^{-1}$	Twice per week laboratory sample	
Carbon (da graphite)	$< 0.05 \text{ mg} \cdot \text{L}^{-1}$	Once a year laboratory sample	
Silver (total)	$< 0.05 \text{ mg} \cdot \text{L}^{-1}$	Monthly laboratory sample	
Copper (total)	$< 0.05 \text{ mg} \cdot \text{L}^{-1}$	Monthly laboratory sample	
Nitrate (NO ₃ -)	$< 0.05 \text{ mg} \cdot \text{L}^{-1}$	Twice per week laboratory sample	
Dissolved substances	$< 0.1 \text{ mg} \cdot \text{L}^{-1}$	Monthly laboratory sample	
Fe (total)	$< 0.1 \text{ mg} \cdot \text{L}^{-1}$	Monthly laboratory sample	
Sulfate (SO ₄ ²⁻)	$< 0.1 \text{ mg} \cdot \text{L}^{-1}$	Twice per week laboratory sample	
Sólids	$< 5 \text{ mg} \cdot \text{L}^{-1}$	Monthly laboratory sample	
Ca, Na, Mg	< 50 ppb (each one)	Monthly laboratory sample	
Al, Zn, Sr, Ba, Pb, Cr, Co	< 50 ppb	Monthly laboratory sample	

 Table 1: IAEA recommended limits and monitoring frequency for the physical-chemical parameters of the primary cooling water of nuclear research reactors (adapted from [4]).

The Figure 1 shows a top view from the reactor's pool and the procedure for collecting water samples.

2.2. Water samples collection and Triga's water chemistry program

The water samples are collected according to the Table 2 by a reactor operator directly from the pool. Any additional procedure is necessary before delivering them to the respective laboratory. All the analyses were carried out by Analytical Chemistry Unit - Classical Techniques Laboratory (UQA-LTC) of the CDTN. Potentiometric and electric conductivity analytical techniques were used

for the measurements of pH and conductively, respectively. In both cases, the samples are analyzed just after the collection. Any additional preparation is necessary. All samples are kept in closed polyethylene bottles [18].

Table 2 shows the updated version of Triga's water chemistry program, adapted from [7,12] is exhibit in. It contains some of the information about the chemical tests performed.

Figure 1: Top view from the reactor's pool, and the core, 6.0 m below the floor level (left). Water samples collection from the pool (right)



Table 2: Simplified version of Triga's water chemistry program

Parameter	Monitoring frequency	Sample volume	Bottle type
Electrical conductivity	Weekly	250 mL	Polyethylene
pH	Weekly	250 mL	Polyethylene
Total alpha e total beta radiation	Weekly	100 mL	Glass
Sulfate, nitrite, nitrate, chloride, and ammonium	Fortnightly	50 mL	Polyethylene
Metals by ICPMS (Al, Zn, Sr, Ba, Pb Cr, Co, Ag, Cu, Ca, Mg, Cs, La, U, Na e Fe)	Monthly	50 mL	Polypropylene

The concentration of sulfate (SO₄⁻²), chloride (Cl⁻), nitrite (NO₂⁻), nitrate (NO₃⁻) and ammonium (NH₄⁺) ions are quantified at the Analytical Chemistry Unit – Liquid Chromatography Laboratory (UQA-LLC), using High Performance Liquid Chromatography (HLPC) method. After collection, the sample must be filtered (pore size: 0.45 μ m), and refrigerated at 4 °C [18].

The assessment of total alpha and beta radiation are performed at the Low Activity Radiochemistry Unit (Urba). The analytical technic considered in this case is Evaporation/Counting proportional. Finally, metals in Table 2 are quantified by ICP-MS (Inductively Coupled Plasma Mass Spectrometry) technic at Analytical Chemistry Unit-Laboratory of Mass Spectrometry (UQA-LEM). The sample is filtered at the time of collection in a membrane (pore size: 0.45 μ m) and acidified with ultrapure HNO₃. Then, it must be shaken and refrigerated at 4 °C [18].

3. RESULTS AND DISCUSSION

3.1 Electrical conductivity

Figure 2 shows electrical conductivity values, ranging from 0.93 μ S·cm⁻¹ to 1.32 μ S·cm⁻¹, with an average error (measurement uncertainty) of \pm 0.1 μ S·cm⁻¹, for water samples collected from August/2022 to May/2023. The behavior observed is like the ones described in the previous reports [8-10, 14-17], from 2012 to 2019.



Figure 2: Electrical conductivity for Triga's cooling water from August/2022 to May/2023

The IAEA recommends that electrical conductivity for the primary cooling system of research reactors be less than 1.0 μ S·cm⁻¹. However, such a limit was never reached by CDTN's research reactor since its water chemistry program was implemented in 2012, which has a typical conductivity value slightly above 1.0 μ S·cm⁻¹. This situation occurs due to specific aspects explained below.

The IPR-R1 Triga reactor is a pool type open to the atmosphere, which can facilitate the presence of microorganisms, insects, leaves, and others in its water. Additionally, Rodrigues [17] had emphasized that there is no very efficient system for cleaning air in the reactor room. Dust of plants, microorganisms and small insects can be found [3]. Such elements have potential to generate decomposition products, like organic acids, capable of increasing the electrical conductivity above the recommended limit. In addition, Rodrigues [17] pointed out that in a safety analysis report carried out by CDTN, 2.0 μ S·cm-1 was established as the upper limit for electrical conductivity of Triga's cooling water based on the operational history of the installation. In this line of reasoning, Ipen's IEA-R1 reactor works with 2.0 μ S·cm-1 as upper limit for electrical conductivity of its pool water, as can be verified in the works carried out by Carvalheira *et al.* [5] and Cegalla *et al.* [19]. Therefore, from electrical conductivity view point, it is concluded that IPR-R1 reactor operates within safe limits.

3.2 pH - Potential of Hydrogen

The pH values referring to Triga's cooling water from August/2022 to May/2023 are shown in Figure 3. The pH ranged from 5.58 to 6.73, with an error (measurement uncertainty) varying from \pm (0.03 to 0.58) in the period considered. In general, this parameter was within the values suggested by the IAEA but exceeded the recommended values (pH = 6.5) on some specific dates, like between September 19th and October 17th of 2022. Possible explanation for this behavior was irradiation processes in the installation during such period. In all cases, pH never exceeded neutrality condition (pH = 7.0), thus never reaching the basic character (pH > 7).

Historically, from 2012 to 2019, as can be seen in previous CDTN technical reports [8-10, 14-17], pH values for Triga's cooling water were always within the limits recommended by the IAEA [4]. However, on some specific dates, the measured values of this parameter exceeded the suggested upper limit but, they never exceeded the situation of neutral pH or the basic condition.



Figure 3: pH values for TRIGA' cooling water from August/2022 to May/2023

3.3 Concentration of sulfate, chloride, nitrite, nitrate and ammonium ions

Table 3 shows the results of the concentration of sulfate (SO4⁻²), chloride (Cl⁻), nitrite (NO²⁻), nitrate (NO³⁻) and ammonium (NH⁴⁺) ions in water samples collected from Triga's pool between September/2022 and May/2023.

The presence of chloride and sulfate ions, dissolved in water, tends to increase its electrical conductivity, thus facilitating the occurrence of electrochemical processes, which increases the probability of corrosion in the metallic materials of IPR-R1 reactor. On the other hand, regarding nitrite (NO²⁻) ion, adequate concentrations of it promotes the formation of a passivation layer in steel and other ferrous metal alloys [4]. This phenomenon reduces the development of corrosive processes in this type of material. Like nitrite, nitrate ions (NO³⁻) in adequate concentrations also act as corrosion inhibitors [20]. Finally, ammonium (NH⁴⁺) ion is normally related to the presence of organic matter in water. Bearing in mind that Triga reactor is a pool type open to the atmosphere, and that there is no efficient air filtration system in its room, its water is in contact with the fauna

and flora contained in the atmospheric air [3], as discussed in Section 3.1, which may influence the presence of NH⁴⁺ ions in its water.

Test reports provided by UQA-LCL show that the concentration of sulfate, chloride, nitrite, and nitrate ions were below the quantification limits (QL) of the experimental method considered, as can be seen in Table 3. They are: $QL = 0.1 \text{ mg} \cdot \text{L}^{-1}$ during the second half of 2022 for such ions; and $QL = 0.06 \text{ mg} \cdot \text{L}^{-1}$ (Cl⁻), $QL = 0.05 \text{ mg} \cdot \text{L}^{-1}$ (NO²⁻), $QL = 0.09 \text{ mg} \cdot \text{L}^{-1}$ (NO³⁻) and $QL = 0.07 \text{ mg} \cdot \text{L}^{-1}$ (SO4⁻²) for the first half of 2023. A similar behavior was observed for ammonium ion, where its concentration was below the quantification limit, in this case 0.05 mg $\cdot \text{L}^{-1}$ for the second half of 2022 and 0.09 mg $\cdot \text{L}^{-1}$ during the first half of 2023.

	ions [mg·L ⁻¹]				
Date	chloride (Cl ⁻)	nitrite (NO ²⁻)	nitrate (NO ³⁻)	sulfate (SO4 ⁻²)	ammonium (NH ⁴⁺)
05/09/2022	< 0.1 (QL)	< 0.1 (QL)	< 0.1 (QL)	< 0.1 (QL)	< 0.05 (QL)
20/09/2022	< 0.1 (QL)	< 0.1 (QL)	< 0.1 (QL)	< 0.1 (QL)	< 0.05 (QL)
03/10/2022	< 0.1 (QL)	< 0.1 (QL)	< 0.1 (QL)	< 0.1 (QL)	< 0.05 (QL)
24/10/2022	< 0.1 (QL)	< 0.1 (QL)	< 0.1 (QL)	< 0.1 (QL)	< 0.05 (QL)
07/11/2022	< 0.1 (QL)	< 0.1 (QL)	< 0.1 (QL)	< 0.1 (QL)	< 0.05 (QL)
21/11/2022	< 0.1 (QL)	< 0.1 (QL)	< 0.1 (QL)	< 0.1 (QL)	< 0.05 (QL)
05/12/2022	< 0.1 (QL)	< 0.1 (QL)	< 0.1 (QL)	< 0.1 (QL)	< 0.05 (QL)
23/01/2023	< 0.06 (QL)	< 0.05 (QL)	< 0.09 (QL)	< 0.07 (QL)	< 0.09 (QL)
27/02/2023	< 0.06 (QL)	< 0.05 (QL)	< 0.09 (QL)	< 0.07 (QL)	< 0.09 (QL)
13/03/2023	< 0.06 (QL)	< 0.05 (QL)	< 0.09 (QL)	< 0.07 (QL)	< 0.09 (QL)
24/04/2023	< 0.06 (QL)	< 0.05 (QL)	< 0.09 (QL)	< 0.07 (QL)	< 0.09 (QL)
08/05/2023	< 0.06 (QL)	< 0.05 (QL)	< 0.09 (QL)	< 0.07 (QL)	< 0.09 (QL)

Table 3: Ions in water samples collected from Triga's pool between September/2022 and May/2023

* QL: Quantification Limit of the experimental method used to carry out the tests.

Regarding the limits of each of these parameters, considering the primary cooling system of research reactors, the IAEA recommends that nitrate and chloride concentrations be below

 $0.05 \text{ mg} \cdot \text{L}^{-1}$, while the limit suggested for sulfate ion is 0.1 mg $\cdot \text{L}^{-1}$, the same value as the quantification limit. In the case of chloride and nitrate ions, the maximum limit recommended by the IAEA is lower than their quantification limit. Thus, there is a possibility that the concentration of such ions (chloride and nitrate) is higher than the recommended one. However, as such ions are usually impurities contained in water, and given that IPR-R1 Triga reactor uses ultrapure H₂O; it contains a low concentration of impurities, combined with an ion exchange system (resin) that adjusts the chemical composition of water. So, the tendency is for such ions to be present in low concentrations that do not compromise the safe operation of the installation [4].

There are no operational limits recommended by the IAEA [4] for nitrite and ammonium ions. However, considering their concentration is below quantification limits, combined with the characteristics of Triga's cooling water (ultrapure and ion exchange resin), the tendency is for these two ions to be present in low concentrations that do not affect IPR-R1 normal performance. Finally, in relation to such parameters (sulfate, chloride, nitrite, nitrate, and ammonium), in general, they always presented historical values (2012 to 2019) below the respective quantification limits, as can be checked in previous CDTN reports [8-10, 14-17], referring to the results of Triga's water chemistry program between 2012 and 2019.

3.4 Alpha and beta radiation

Figures 4 and 5 exhibit the total values of alpha (α) and beta (β) radiation for the water samples collected in TRIGA's pool between August/2022 and June/2023. The measurement of α and β can be used as an indicative of the presence of fission products in its cooling water, both arising from the activation of samples inserted into reactor core, and those arising from the rupture of fuel elements [7].

In the case of alpha, it ranged from 0.02 $Bq\cdot kg^{-1}$ to 0.48 $Bq\cdot kg^{-1}$, with an average error (measurement uncertainty) of \pm 0.05 $Bq\cdot kg^{-1}$; while beta ranged from 2.3 $Bq\cdot kg^{-1}$ to 30 $Bq\cdot kg^{-1}$ with an average error (measurement uncertainty) of \pm 0.2 $Bq\cdot kg^{-1}$. About the minimum and maximum values of α and β recommended for the cooling system of research reactors, there is no explicit recommendation in IAEA reference document [4]. However, the values of alpha and beta in Figures 4 and 5 are below the values considered acceptable for the installation, having as reference

the detected values for both parameters in previous Triga's water chemistry program reports [8-10, 14-17], considering the years 2012 to 2019. By comparison, according to program results during 2012 [11] [8], alpha ranged from approximately 0 to 1 Bq·kg⁻¹ while beta ranged from 0 to 200 Bq·kg⁻¹. In this situation, the technical team responsible for preparing such report [8] concluded that those values were within the acceptable range considering IPR-R1 operational routine in that period. Then, in the context of alpha and beta radiation, can be considered that IPR-R1 research reactor operates in a safe way.





Figure 5: Total beta (β) radiation for Triga's cooling water from August/2022 to June/2023



3.5 Metals detected by ICP-MS

Table 4 and 5 show the measured values of metals, detected by ICP-MS technique, in Triga's water samples considering the second half of 2022 and the first half of 2023 including the values of QL (Quantification Limit).

Element	Concentration (μg·L ⁻¹) 22/08/2022	Concentration (µg·L ⁻¹) 05/09/2022	Concentration (μg·L ⁻¹) 03/10/2022	Concentration (µg·L ⁻¹) 07/11/2022	IAEA limits (µg·L ⁻¹) [4]
Li	< 0.1 (QL)	< 0.1 (QL)	< 0.1 (QL)	< 0.1 (QL)	
Cu	< 0.7 (QL)	< 0.7 (QL)	< 0.7 (QL)	< 0.7 (QL)	< 50
Sr	< 1 (QL)	< 1 (QL)	< 1 (QL)	< 1 (QL)	< 50
Ag	< 0.005 (QL)	< 0.005 (QL)	< 0.005 (QL)	< 0.005 (QL)	< 50
Cs	< 0.005 (QL)	< 0.005 (QL)	< 0.005 (QL)	< 0.005 (QL)	
U	< 0.005 (QL)	< 0.005 (QL)	< 0.005 (QL)	< 0.005 (QL)	
Na	< 7 (QL)	< 7 (QL)	< 7 (QL)	< 7 (QL)	< 50
Al	< 10 (QL)	< 10 (QL)	< 10 (QL)	< 10 (QL)	< 50
Mn	< 1 (QL)	< 1 (QL)	< 1 (QL)	< 1 (QL)	
Fe	< 10 (QL)	< 10 (QL)	< 10 (QL)	< 10 (QL)	< 100
Zn	4 (QL = 3)	6.2 (QL = 3)	3.1 (QL = 3)	< 3 (QL)	< 50
La	< 0.01 (QL)	< 0.01 (QL)	< 0.01 (QL)	< 0.01 (QL)	

Table 4: Metals detected by ICP-MS during the second half of 2022

Table 5: Metals detected by ICP-MS during first half of 2023

Element	Concentration (μg·L ⁻¹) 19/12/2022	Concentration (μg·L ⁻¹) 23/01/2023	Concentration (μg·L ⁻¹) 27/02/2023	Concentration (µg·L ⁻¹) 27/03/2023	IAEA limits (μg·L ⁻¹) [4]
Li	< 0.1 (QL)	< 0.1 (QL)	< 0.1 (QL)	0.5 (QL)	
Cu	< 0.7 (QL)	< 0.7 (QL)	< 0.7 (QL)	3.76 (QL < 0,3)	< 50
Sr	< 1 (QL)	< 1 (QL)	< 1 (QL)	0.42 (QL < 0,2)	< 50
Ag	< 0.05 (QL)	< 0.05 (QL)	< 0.05 (QL)	< 0.2 (QL)	< 50
Cs	< 0.05 (QL)	< 0.05 (QL)	< 0.05 (QL)	< 0.2 (QL)	
U	< 0.05 (QL)	< 0.05 (QL)	< 0.05 (QL)	< 0.2 (QL)	
Na	< 7 (QL)	< 7 (QL)	< 7 (QL)	< 13 (QL)	< 50
Al	< 10 (QL)	< 10 (QL)	< 10 (QL)	< 8 (QL)	< 50
Mn	< 1 (QL)	< 1 (QL)	< 1 (QL)	< 0.4 (QL)	
Fe	< 10 (QL)	< 10 (QL)	< 10 (QL)	< 26 (QL)	< 100
Zn	3 (QL = 3)	3(QL = 3)	3.1 (QL = 3)	4.36 (QL = 3)	< 50
La	< 0.01 (QL)	< 0.01 (QL)	< 0.01 (QL)	< 0.05 (QL)	

In general, the results presented in both tables are below the quantification limits of the experimental method used to carry out the tests, and below the limits recommended by the IAEA [4], as can be compared with the reference values in Table 1. Besides that, such results agree with the global behavior of previous CDTN reports [8-10, 14-17] covering the results (metals detected by ICP-MS) of Triga's water chemistry program between 2011 and 2019. The results exhibited in Tables 4 and 5, confirms that the concentration of metals of Triga's water agree with the recommended parameters.

4. CONCLUSIONS

In this paper, the results referring to the water quality management program for the cooling water of IPR-R1 Triga research reactor for the period between August/2022 and June/2023, when the installation returns to its regular activities after the Covid-19 pandemic, were presented and discussed.

In general, the results for the parameters evaluated, pH, electrical conductivity, and ions (sulfate, nitrite, nitrate, chloride, and ammonium) were within the limits recommended by the IAEA or they were within the values considered normal (or acceptable) by CDTN technical team due to the operational history of the facility.

Regarding the parameters total alpha and total beta, although such parameters do not have operational limits recommended (defined) by the IAEA, the values presented for the period in question (2022-2023) are within the range considered normal based on previous internal CDTN reports prepared by the technical team responsible for Triga's water quality management program in previous years (2012 to 2019). The development of a corrosion monitoring program for Triga research reactor, in order to complement its water chemistry program is under way.

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