

WATER CHEMICAL CONTROL OF THE TRIGA IPR-R1 REACTOR PRIMARY COOLING SYSTEM

Lucia M. L. A. Auler, Renata D. A. Chaves, Helena E. L. Palmieri, Maria Ângela de B. C. Menezes, Paulo F. Oliveira, Geraldo F. Kastner, Ilza Dalmázio, Oliene dos R. Fagundes, Maria Olivia C. Cintra, Geraldo V. de Andrade, Ângela M. Amaral, Milton B. Franco, Flávio Fortes, Nilton Carlos Gomes, Andréa Vidal, Fausto Maretti Junior, Eliana A. N. Knupp, Wagner de Souza, João B. Guedes, Renato C. S. Furtado

Centro de Desenvolvimento da Tecnologia Nuclear (CDTN / CNEN)
Av. Pres. Antônio Carlos, 6.627 - Campus UFMG, CEP 31270-901 - Caixa Postal 941, CEP 30161-970
Belo Horizonte, Minas Gerais

aulerlm@cdtn.br, rda@cdtn.br, help@cdtn.br, menezes@cdtn.br, pfo@cdtn.br, gfk@cdtn.br, id@cdtn.br, orf@cdtn.br, mocc@cdtn.br, gva@cdtn.br, ama@cdtn.br, francom@cdtn.br, fortes@cdtn.br, ncg@cdtn.br, avf@cdtn.br, fmj@cdtn.br, ean@cdtn.br, wagner@cdtn.br, jbg@cdtn.br, furtado@cdtn.br

ABSTRACT

The TRIGA MARK I IPR-R1 reactor located at CDTN/CNEN has been in operation and contributed to research and with services to society since 1960. It has been used in several activities such as nuclear power plant operation, graduate and post-graduate training courses, isotope production, and as an analytical irradiation tool of different types of samples. Among the several structural and operational safety requirements is the chemical quality control of the primary circuit cooling water. The aim of this work was to check the cooling water quality from the pool reactor. A water sampling plan was proposed (May, 2011 – June, 2012) and presents the results obtained in this period. The natural radioactivity level as gross alpha and gross beta activity and other chemical parameters (pH and electric conductivity) of the samples were analyzed. Some instrumental techniques were used: potentiometric methods (pH), conductometric methods (electrical conductivity, EC) and gross α and gross β (proportional counting system).

1. INTRODUCTION

The TRIGA MARK I IPR-R1 Reactor, located at Centro de Desenvolvimento da Tecnologia Nuclear/Comissão Nacional de Energia Nuclear, CDTN/CNEN, runs at 100 kW. The reactor uses 20% enriched uranium as fuel in a homogeneous alloy with zirconium hydrate as the main moderator. The reactor core is illustrated in Fig. 1.

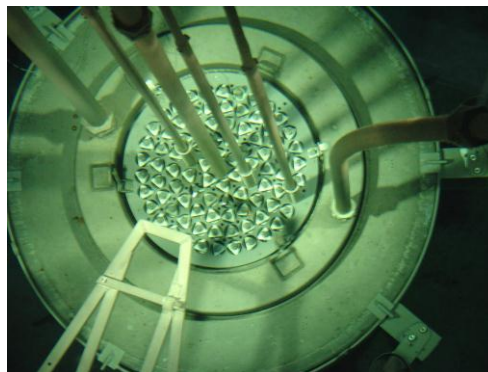


Figure 1: TRIGA MARK I -IPR-R1 Nuclear reactor, CDTN/CNEN

The cooling water, which is light and demineralized, serves as an additional moderator and a biological shield. Aging is defined as the general process by which the component characteristics gradually change over time or with use under normal operation conditions with their resulting degradation [1]. An Aging Management Program has already been defined for the TRIGA IPR-R1 reactor. It details the aging mechanisms and effects under normal operation conditions.

There is a standard for research reactors that uses a mixture of fuel elements clad with different elements such as aluminum and stainless steel. It establishes the analytes that most contribute to the corrosive process as well as the recommended sampling and minimal limits [2]. A corrosion study of aluminum alloys exposed to the TRIGA IPR-R1 reactor has also been carried out. This evaluation was carried out under the Regional Technical Cooperation (Project Regional de Cooperação Técnica) RLA/4/018 funded by IAEA [3].

The chemical conditions maintained at different pool reactors are usually very similar. Ion exchange resins are used to maintain high water purity in the pool. Conductivity is the main indicator of water purity. Naturally, corrosion of aluminum alloys in the reactor coolant may be minimized by the careful control of water purity, which is an important safety requirement.

A variety of analytical chemical tasks is needed to support reactor operation. Routine analysis should be conducted to check the operational condition of the reactor. A large percentage of analytical problems in the reactor is related to the determination of either fission products and impurity trace elements that may result from the introduction of either experimental devices or manipulation tools. Radiochemical methods are often highly sensitive to such changes. Furthermore, the value of conventional methods of analysis, such as spectroscopy, colorimetry and some separation techniques should not be underestimated [4].

Determinations have been carried out at several sampling points to meet the quality control requirements for the reactor cooling system water. It is worth noting that this control has been resumed after a new TRIGA IPR-R1 reactor configuration was established, particularly with the inclusion of stainless steel-clad fuel elements.

The reactor deionized water is cooled by a stainless steel shell-and-tube-type heat exchanger in a room next to the reactor. In the primary circuit, water goes through the shield at 28 m³/h and in the secondary circuit it passes through the tubes. The primary circuit water is pumped from the bottom of the reactor pool through the stainless steel tubes by a mechanically sealed pump to the heat exchanger, where it is circulated and returned to the pool at mid-height, providing constant homogenization. The secondary circuit water (without treatment) is cooled in an external tower (air/water).

Reactor water purification is done by a mixed-bed resin system in a steel tank coated with epoxy and lined with two 50- μ m fiber filters (tank inlet and outlet) to retain suspended solid particles. Water is circulated through the resins once a month, which allows keeping conductivity between 1.25 and 1.45 μ S.cm⁻¹. An aluminum tank of approximately 250 L capacity holds reserve distilled water used to compensate water loss by evaporation [3].

2. REACTOR WATER MONITORING PROGRAM

Pool surface and bottom points are important for monitoring chemical species in the reactor water. At first, sampling at mid-depth is not considered necessary as the water flow distribution inside the pool is homogeneous. Table 1 presents the chemical control program for the TRIGA reactor located at CDTN.

Table 1. Analysis of the water of the TRIGA MARK I IPR-R1 reactor

Parameters	Analytical Techniques	Frequency
Conductivity	Direct Reading	Weekly
pH	Direct Reading	Weekly
Gross alpha and Gross beta	Radiometry	Weekly
Gamma	Espectrometry	Weekly
B, Na, Fe, Si	Atomic absorption spectrometry and Inductive coupled plasma (ICP-AES)	Fortnightly
Ag, Al, Cs, Cu, Li, Mn Sr, U and Zn	Atomic absorption spectrometry and Inductive coupled plasma (ICP –MS)	Monthly
Hg	Cold vapor atomic absorption spectrometry (CVAAS)	Three-monthly
ammonium ion	Ionic Exchange Chromatography	Fortnightly
sulphate, nitrate, nitrite and chloride ion	Ionic Exchange Chromatography	Fortnightly

3. RESULTS AND DISCUSSION

3.1. Conductivity and pH

Figure 1 shows the analysis results of conductivity and pH, respectively, in this period. The measurement of the conductivity of the water pool TRIGA reactor should be maintained between 1.0 and 3.0 $\mu\text{S}\cdot\text{cm}^{-1}$ and pH between 5.5 to 7.0 considering the pH range of passivated aluminum. It was observed that the results for both parameters are within appropriate ranges.

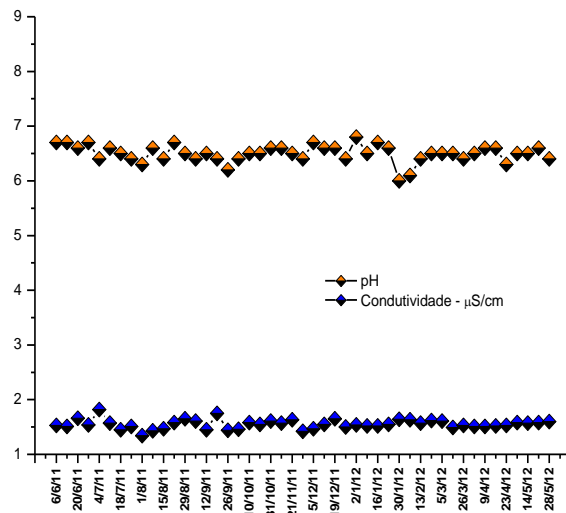


Figure 1: Evolution of pH and conductivity ($\mu\text{S}\cdot\text{cm}^{-1}$) in the reactor from June 2011 to May 2012.

3.2. Gross alpha and Gross beta

Alpha activity present in reactor water should always be near the detection limit of the technique, since the presence of alpha emitters may be indicative of a fault in the fuel element. Beta activity, in turn, may indicate the activation of corrosion products or other elements from the environment, as well as the presence of fission products. Beta activity can vary correlated reactor operations. Activities in water are higher after long irradiations.[5,6,7,8]

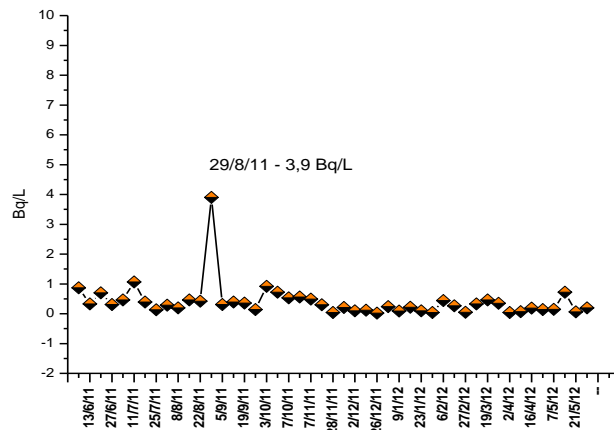


Figure 2: Specific Gross alpha activity in the reactor from June 2011 to May 2012

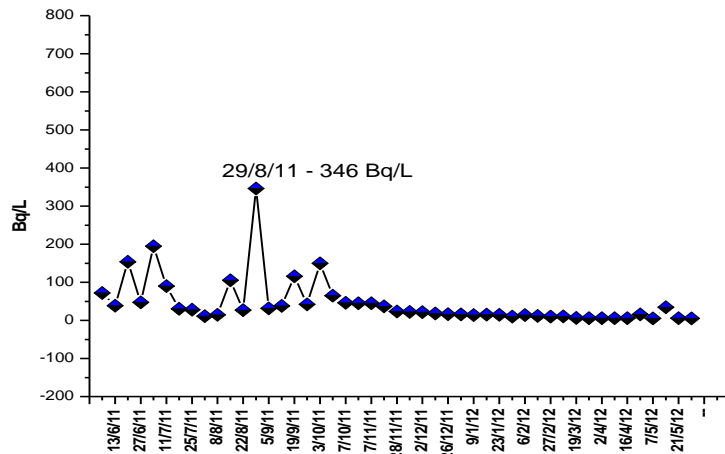


Figure 3. Specific Gross beta activity in the reactor from June 2011 to May 2012

According to Figures 2 and 3, the presence of "outlier" points was observed (August 29, 2011), whose behavior was investigated although not explained. The results for gross alpha and gross beta activities are similar to those reported in previous studies [5,6]. The latter showed similar activity ranges: 0.05 to 1.50 Bq. L⁻¹ for alpha and 5 – 200 Bq. L⁻¹ for beta, including the presence of "outliers". In Figure 3, there is a region at the beginning of the graph corresponding to higher values of beta activity registered after long irradiation. This region, from June to October 2011, was a period of intense reactor operation.

3.3. Gamma Spectrometry

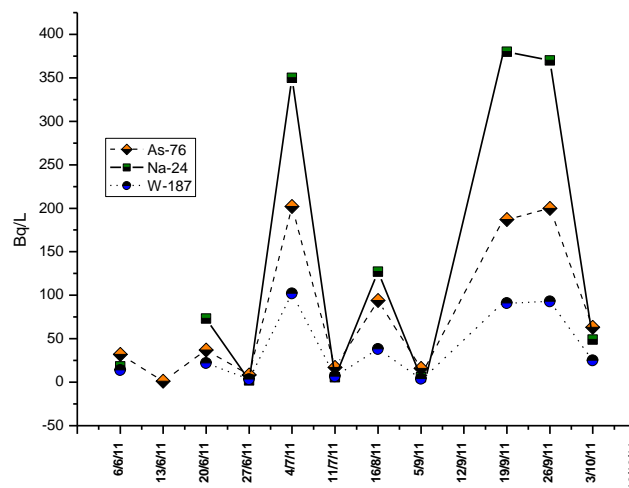


Figure 4: Specific activity of gamma emitters (⁷⁶As, ²⁴Na and ¹⁸⁷W) in the reactor from June 2011 to May 2012.

According to Figure 4, the presence of ^{24}Na can be explained by the reaction of the aluminum structure of the reactor with fast neutrons $^{27}\text{Al}(n_f, \alpha)^{24}\text{Na}$ or by sodium activation (isotopic abundance 100% in the environment), according to the reaction $^{23}\text{Na}(n_f, \gamma)^{24}\text{Na}$. Other radionuclides such as ^{58}Co and ^{54}Mn when detected, may come from reactions of other nuclides with fast neutrons like $^{54}\text{Fe}(n_f, p)^{54}\text{Mn}$ and $^{58}\text{Ni}(n_f, p)^{58}\text{Co}$. It can happen because iron is also a component of environmental dust and nickel is part of the structure of the reactor. Other radionuclide, ^{76}As , comes from the dust in the air, while ^{60}Co , ^{65}Zn and ^{187}W may come from the dust of the environment and from corrosion processes [3]. This provenance should be investigated during the control program.

3.4. Ag, Al, Cs, Cu, Li, Mn, Sr, U and Zn concentrations

Table 3: Li, Al, Mn, Cu and Zn concentrations in the water reactor from June 2011 to May 2012

Data	Li	Al	Mn	Cu	Zn
	($\mu\text{g/L}$)	($\mu\text{g/L}$)	($\mu\text{g/L}$)	($\mu\text{g/L}$)	($\mu\text{g/L}$)
06/June/2011 to 01/May/2012	< 0.1- < 0.3	< 0.2 - 9.94	< 0.4 – 3.4	< 0.34– 1.43	4.81 - 112

According to Howell [5], the levels found are considered suitable for protection coatings of aluminum alloys against corrosion.

3.5. Hg concentration

Mercury concentration was determined by Atomic Absorption Spectrometry with Cold Vapor Operating Procedure - PO LAA 03 Rev. 06 [9]. All determinations gave results below the detection limit of the technique ($0.1 \mu\text{g L}^{-1}$).

3.6. Ammonium, sulphate, nitrate, nitrite and chloride ions concentrations

Concentrations values obtained for the ions in the samples collected in this period (2011-2012) were below the detection limit of the methodology available at CDTN. The anions results were $<0.25 \text{ mg. L}^{-1}$ and the ammonium ion was $<0.1 \text{ mg. L}^{-1}$ corresponding to the detection limit of ion-exchange chromatography, specific to these analytes.

4. CONCLUSIONS

There is little information regarding a TRIGA type reactor monitoring program in the literature. The main objective of this program is to prevent, minimize and / or control the corrosive processes arising from the various chemical reactions that can be produced in the reactor structure. The analysis was carried out during an one - year period and the results of chemical and radiochemical analyses indicated that TRIGA MARK I IPR-R1 water reactor did not show significant concentrations of ions as observed by monitoring measurements. The presence of radionuclides that may lead to loss of fuel, was not detected. The conclusions

were made considering that monitoring conductivity is a tool to evaluate the ion concentration in the pool water reactor.

Finally, it was concluded that this program is an important tool for monitoring reactor water and the information resulting from this monitoring will strongly contribute to similar programs applied to other research reactors.

5. REFERENCES

1. J.R.B. Cruz, Z. Rocha, A. Z. Mesquita, P.F”. Oliveira, R.R. Rodrigues, Programa de Gestão de Envelhecimento para o Reator TRIGA IPR-R1”. In: XII Encontro Nacional de Física de Reatores e Termohidráulica, Rio de Janeiro. ABEN, 2000. v. CD. (2000).
2. Standard Guidelines for Corrosion Protection of Research Reactor Aluminum-Clad Spent Nuclear Fuel in Interim Wet Storage (Draft International Standard ISSO/TCxxx/SC VER. 6).
3. C. F. C. Neves, P. F. Oliveira, W. R. C. Campos, J. E. Campos F°, “Corrosão de ligas de alumínio no reator TRIGA IPR-R1”, Belo Horizonte: Centro de Desenvolvimento da Tecnologia Nuclear / CNEN, , 19p. (Relatório final de projeto), Nº RD-EC2-001/06 (2006).
4. International Atomic Energy Agency, Research Reactor Utilization, Technical Reports Series Nº 71, Vienna, 89 p, (1967).
5. J. P. Howell. Criteria for corrosion protection of alluminium-clad spent nuclear fuel in interim wet storage. In: CORROSION, 2000, march 26 – 31 Orlando, Flórida. Proceedings. Houston: NACE, 2000. Paper nº 200, 2000.
6. C. V. S Sabino. Controle Químico da água de refrigeração do reator TRIGA IPR-R1 (05/98 a 04/99). Belo Horizonte: CDTN, 1999. (Nota Interna)
7. C. V. S Sabino, P. F. Oliveira, Controle Químico da água de refrigeração do reator TRIGA IPR-R1 (05/99 a 07/2000). Belo Horizonte: CDTN, 2000. (Nota Interna)
8. C. V. S Sabino, N. Bazzoli, A. M. Amaral. **Técnicas Analíticas Nucleares no controle da qualidade de águas**, Belo Horizonte: CDTN, 1996. 6p.
9. Procedimento Operacional – PO LAA 03 Rev. 06.