SYSTEM TO DETECT LEAKING IN FUEL ELEMENTS IN TRIGA NUCLEAR RESEARCH REACTOR

SISTEMA PARA DETEÇÃO DE VAZAMENTOS EM ELEMENTOS COMBUSTÍVEIS NOS REATORES NUCLEARES DE PESQUISA TRIGA

A. Z. Mesquita[1], R. R. Rodrigues[2]

[1]Pesquisador Titular, CDTN/Cnen, Belo Horizonte - Brasil.
[2]Tecnologista Senior, CDTN/Cnen, Belo Horizonte - Brasil..

ABSTRACT

The Nuclear Technology Development Center (CDTN) is a research institute administered by the Brazilian Nuclear Energy Commission (CNEN) located in Belo Horizonte/Brazil). The CDTN is equipped with a Triga Mark I nuclear reactor with almost 60 years of operation. Most of its fuel elements are in the core since the first criticality, and corrosion may occur that threaten the integrity of fuel claddings. The reactor coolant must be treated and controlled in order to maintain its low electrical conductivity and pH close to neutrality, in order to minimize the corrosion of the reactor components, mainly the fuel elements. The objective of this work is to present the leak monitoring system developed for the verification of possible leaks in the Triga fuel elements based on the sipping test. A correlation was also developed to find the diameter of a hypothetical small cylindrical hole in the fuel cladding as a function of Cs-137 activity values to be measured.

RESUMO

O Centro de Desenvolvimento de Tecnologia Nuclear (CDTN) é um instituto de pesquisa administrado pela Comissão Nacional de Energia Nuclear (Cnen), localizada em Belo Horizonte/Brasil. O CDTN está equipado com um reator nuclear Triga Mark I com quase 60 anos de operação. A maioria de seus elementos combustíveis está no núcleo desde a primeira criticalidade, e pode ocorrer corrosão que ameaça a integridade dos revestimentos de combustível. O refrigerante do reator deve ser tratado e controlado, a fim de manter sua baixa condutividade elétrica e pH próximo à neutralidade, a fim de minimizar a corrosão dos componentes do reator, principalmente, os elementos combustíveis. O objetivo deste trabalho é apresentar o sistema de monitoramento desenvolvido para a verificação de possíveis vazamentos nos elementos combustíveis Triga baseados no teste de sipping. Também foi desenvolvida uma correlação para encontrar o diâmetro de um pequeno buraco cilíndrico hipotético no revestimento de combustível como uma função dos valores de atividade do Cs-137 a serem medidos.

1. INTRODUCTION

Triga reactors are the most widely used research reactor in the world. There is an installed base of over sixty-five facilities in twenty-four countries on five continents. General Atomics (GA), the supplier of Triga research reactors, since the late 1950s continues to design and install Triga reactors around the world and has built Triga reactors in a variety of configurations and capabilities, with steady state thermal power levels ranging from 100 kW to 16 MW.

Triga reactors are used in many diverse applications, including production of radioisotopes for medicine and industry, treatment of tumors, nondestructive testing, basic research on the properties of matter, and for education and training. The Triga reactor is the only nuclear reactor in this category that offers true "inherent safety," rather than relying on "engineered safety." It is possible due to the unique properties of GA's uranium-zirconium hydride fuel, which provides unrivaled safety characteristics, which also permit flexibility in sitting, with minimal environmental effects [1].

The IPR-R1 Triga (Instituto de Pesquisas Radiativas - Reactor 1, Training Research Isotope General Atomic) reactor, located at the Nuclear Technology Development Center - CDTN (Belo Horizonte/Brazil), is a typical Triga Mark I light-water and open-pool type reactor. The fuel elements in the reactor core are cooled by water natural circulation. The heat removal capability of this process is great enough for safety reasons at the current maximum 250 kW power level configuration.

The IPR-R1 reached its first criticality on November 1960 with a core configuration containing 56 aluminum clad standard Triga fuel elements, and a maximum thermal power of 30 kW. In order to upgrade the IPR-R1 reactor power, nine stainless steel clad fuel elements were purchased in 1971. One of these fuel elements was instrumented in the centerline with three type K thermocouples. On December 2000, four of these stainless steel clad fuel elements were placed into the core allowing to upgrading the nominal power to 250 kW. In 2004 the instrumented fuel element was inserted into the core and monitored the fuel temperature [2]. The basic safety limit for the Triga reactor system is the fuel temperature, both in steady-state and pulse mode operation.

The IPR-R1 Triga core (Figure 1) is placed at the bottom of an open tank of about 6 m height and 2 m diameter. The tank is filled with approximately 18 m2 of water able to assure an adequate radioactive shielding. The core has an annular graphite reflector and cylindrical configuration of six rings with 90 positions able to host either fuel rods or other components like control rods, graphite dummies (mobile reflector), irradiating and measurement channels.

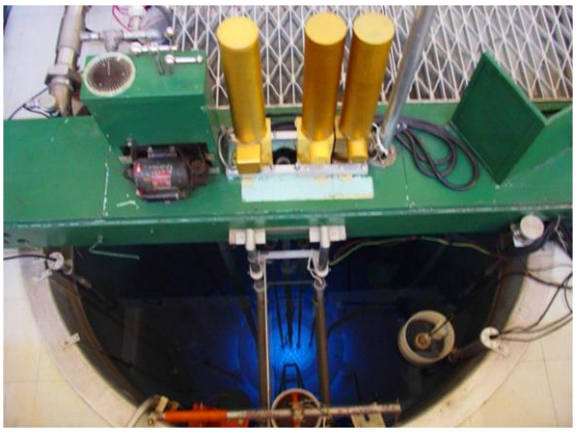

Fig. 1 - The IPR-R1 Triga nuclear reactor [2].

A simplified diagram of the IPR-R1 Triga reactor is show in Figure 2. The prototypical cylindrical fuel elements are a homogeneous alloy of zirconium hydride (neutron moderator) and uranium enriched at 20% in 235U (Fig. 3). The reactor core has 58 aluminum-clad fuel elements and five stainless steel-clad fuel elements. The fuel rod has about 3.5 cm diameter; the active length is about 37 cm closed by graphite slugs at the top and bottom ends, which act as axial reflector. The power axial distribution factor in the fuel is 1.25 [3]. The moderating effects are carried out mainly by the zirconium hydride in the mixture and on a smaller scale by light water coolant. The characteristic of the fuel elements gives a very high negative prompt temperature coefficient and is the main reason of the high inherent safety behavior of the Triga reactors [4]. The power level of the reactor is controlled with three independent control rods.

In the IPR-R1 Triga reactor the 59 original aluminum-clad fuel elements have the wall thickness of 0.76 mm. The five stainless steel-clad fuel elements have a wall thickness of 0.50 mm [5]. These wall thicknesses are designed to be as thin as possible in order to improve the heat conduction and the neutron flux. But, on the other hand, it makes delicate the fuel elements manipulation. The stainless

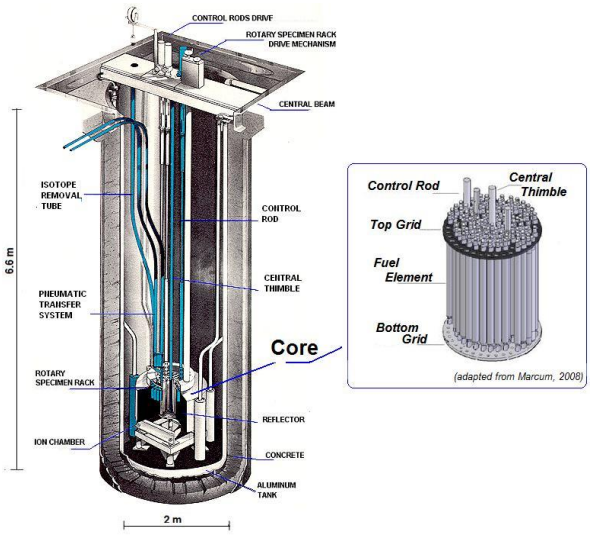

Fig. 2 - IPR-R1 TRIGA reactor pool and core [2].

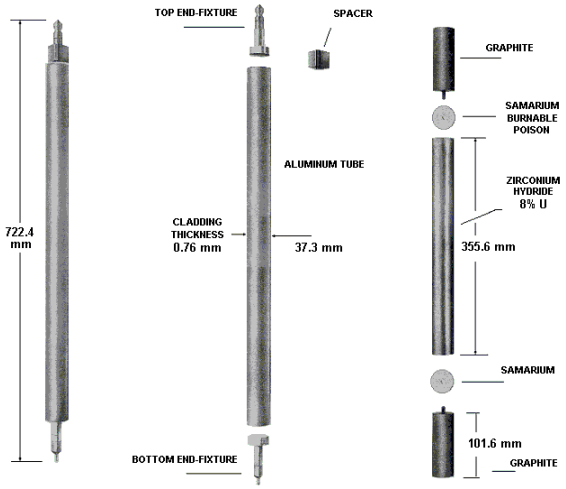

Fig. 3 - Constructive details of a Triga aluminum-clad fuel element [9].

steel-clad fuel elements are mechanically more resistant than the aluminum-clad fuel elements due to the different properties of these materials [6] [7].

During the normal operation of a nuclear reactor, fission product gases accumulate in the fuel-to-clad gap as the uranium dioxide fuel pellets experience nuclear fission. When a fuel clad fails during normal reactor operations, coolant can enter the fuel-to-clad gap, releasing fission products such as volatile species of noble gases or iodine into the primary cooling system [8].

The IPR-R1 Triga is a reactor with almost 60 years of operation. It has been widely used in reactor operator training. In the practical classes the manipulation of the fuel elements occurs, for example, in the subcritical approximation classes. Handling causes cladding friction and can lead to damages such as scratches and holes. In the visual inspection performed by Alencar *et al.* [10] using a submerged camera (Fig. 4) some corrosion pits were observed on some aluminum-clad fuel elements. Other fuel elements had defect on the bottom end-fixture (Fig. 5).

With the modifications introduced into the reactor over the years of operation, especially the inclusion in the core of stainless steel-clad fuel elements. The main reason for the pitting corrosion is the galvanic pair formed between different metals in the same body of electrolyte dispersion [11].

Although the electrolyte diffusion in the reactor pool water is minimal, there is always the incorporation of air dust in the water, causing the presence of dissolved materials. The pool of research reactors is almost open to environment. Therefore, the possibility exists of the presence of foreign matter in the water with the possibility of fuel channel blockage. This event has different characteristics depending on the flow direction. Downward cooling flow can lead to blockage due to objects dropping into the pool. Upward cooling flow can lead to blockage due to objects inside the primary cooling system piping being dragged into the core by the action of the pump [12]. Reis *et al.* [13] performed a theoretical evaluation of the consequences of this accident for the IPR-R1 Triga reactor.

Fuel channel blockage incidents were reported by the IAEA [14] in research reactors. Two events identified that debris of inappropriate material used in the cooling water circuit caused water flow blockages. One of these events occurred in the 1970s in which the primary coolant hold-up tank inner surface rubber liner got separated from the tank. Pieces of the rubber liner blocked the primary circuit pump inlet and caused a flow reduction. The other event from the 1990s reported a blockage of the continuous sampling circuit due to debris of plastic lining of the primary cooling circuit.

Oxidizing gases such as the dissolved oxygen itself in the pool water can increase the concentration of dissolved ions in the coolant.

There is a study conducted by Sabino [15] which report that most of the floating material in pool water is made up of microorganisms. Among these organisms it can mention *Tricoderma sp.* Residues of the water purification resin itself were also present. Precipitates can be viewed on the pool wall (Fig. 6). Mitigating measures were taken to avoid the presence of resin particles in pool water.

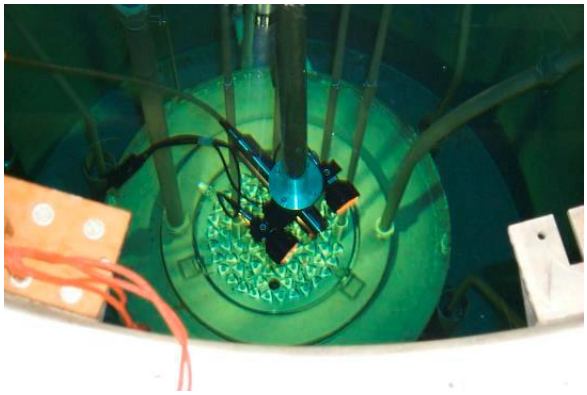

Fig. 4 - Camera immersed in the pool for visual inspection [10].

However, the presence of microorganisms remains and there is a possibility of acidification of water by microbiological activity due to the production and release of organic acids, caused by the microorganism *Trichoderma sp* [15], [16], [17]. These organic acids, which although generally have a high acid dissociation constant in water (pKa), can lead to attacks, especially on aluminum and stainless steel components. These metals are exactly the components of the structures found in the IPR-R1 Triga reactor. This condition may favor the formation of galvanic cells that can lead to electrochemical corrosion in the various structures of the reactor, including the cladding of some of the core fuel elements [11].

A sample of material collected in the water filter was withdrawn from the reactor pool. The material was dissolved in deionized water and seen at a microscope. The images made, from the optical microscopy system, show elements in suspension.

Figure 7 shows three distinct regions with peculiar behaviors. The region marked with a yellow circle behaves like a precipitate of inactive microorganisms. The blue circle labeled region shows smaller particles of clumps

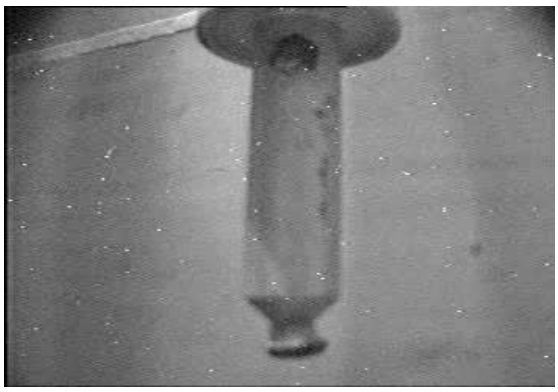

Fig. 5 - Aluminum-clad fuel element with kneaded bottom end-pin [10].

of inactive microorganisms released in the dissolution water stream. The region marked in red shows moving particles of microorganism that apparently manifest their own movement, different from the common movement caused by the Brownian effect observed in the other regions. In the region in red, possibly, it registers biological elements with characteristics of protozoans in process of resuscitation, after contact with the water of dissolution. These results indicate that further investigations should be conducted so that the species present in the coolant are identified and studied from the biological point of view.

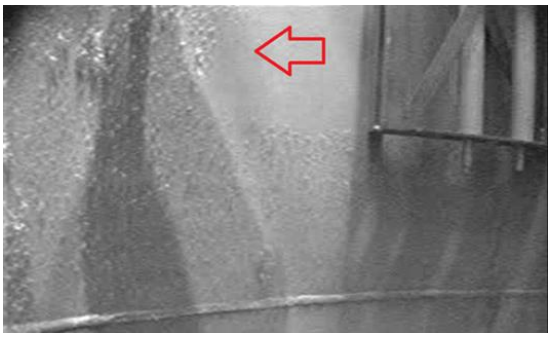

Fig. 6 - Precipitates deposited on the inner wall of the tank [18].

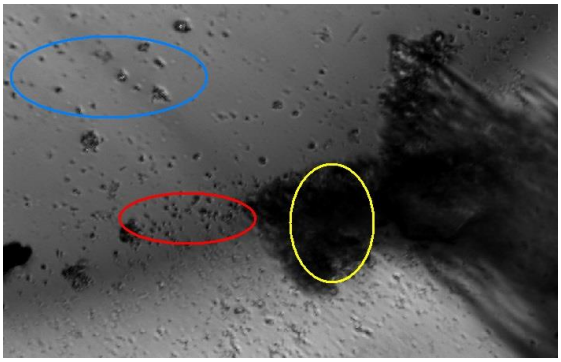

Fig. 7. - Material collected in the coolant filter with magnification of 400x [18].

2. METHODOLOGY

Sipping is the most common technique used to locate fuel failures in both Pressurized Water Reactors (PWRs) and Boiling Water Reactors (BWRs). Identification of fuel rod failure is based on detecting the activity of fission products released through defects during sipping. The measured radioisotopes include krypton, xenon, cesium, and iodine [8].

Sipping is a non-destructive method used to test for failed reactor fuel elements by investigating the fission product released in a fixed volume of core cooling water. This method is usually implemented as a way to monitor to the integrity of fuel elements in research nuclear reactors that can be affected by corrosion over long periods. Sipping can also be employed to investigate fuel elements, which are suspected to have failed during the course of operation [19].

A device was constructed to receive a small number of fuel elements (at most three per test). The handling of the fuel elements is performed through a tool known as "Articulated Clamp for Handling Devices and Remote Equipment", a patented registered by CDTN/Cnen [19]. The device has a vessel filled with deionized water separated from the water in the remainder of the reactor pool. After about 16 hours of fuel elements maceration in the water device container, the water will be sampled. Aliquots of the leaching water will be taken for analysis. Water samples will be analyzed by gamma spectrometry detection using HPGe (High Pure Germanium Spectrometer Multichannel Analyzer).

The analysis will detect and identify fission products that are released in the water sample. The presence of the isotopes: Cs-137, La-140 and I-131, indicates that some of the fuel elements tested have leak, that is, they are compromised. These radioisotopes are indicative of leakage in fuel elements from nuclear reactors. The system will be adapted and will be available to analyze and identify the presence of chemical elements in the coolant in periodic tests.

3. TESTS AND RESULTS

3.1. Sipping device

A device in which fuel elements are to be inserted was designed and constructed as shown in Figure 8. The device consists of an aluminum container with a capacity of about 40 liters of water. Inside the container it is inserted a basket with capacity to be placed up to three fuel elements per test. The basket can be placed outside the pool or positioned on the side of the reactor core so that the operator can transport EC from the core to the basket and vice versa.

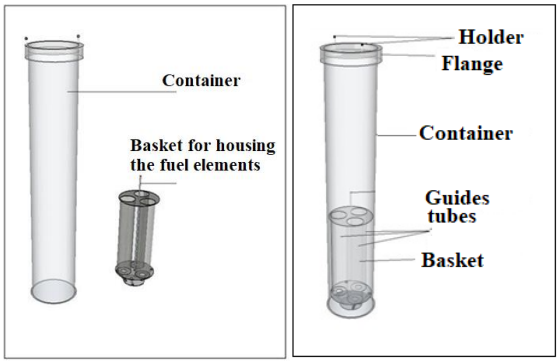

Fig. 8 - Simplified schematic of the sipping apparatus: a) sipping container and basket; b) sipping basket inside the container.

The photographs in Figure 9 show details of the apparatus constructed for carrying out the sipping test. In the photograph shown in Fig. 9a there is the container where the basket with the fuel elements will be inserted. The photo of Figure 9b has the basket with three fuel elements. Figure 9c and Fig. 9d shows detail of the upper and lower terminals of the fuel elements.

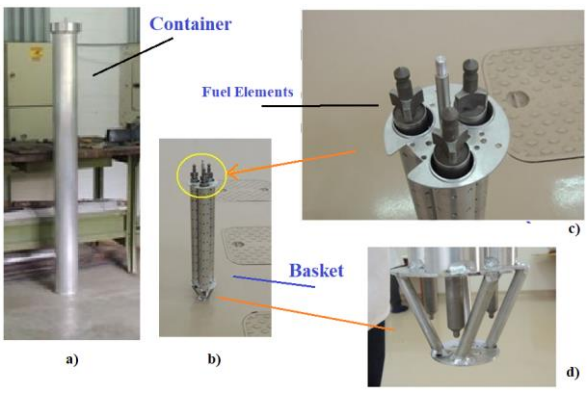

Fig. 9 - Details of the apparatus for sipping test built.

Figure 10 shown the container of the sipping system at the reactor pool bottom to receive fuel elements for the test.

Figure 11 shown the container positioning at the pool edge loaded with three fuel elements submitted to the sipping test.

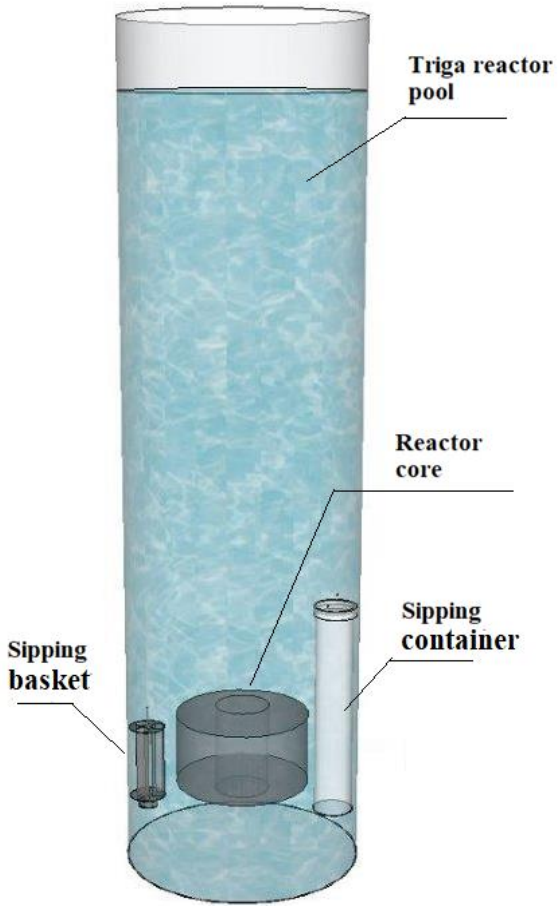

Fig. 10 - Container of the sipping system at the reactor pool bottom to receive fuel elements for the test.

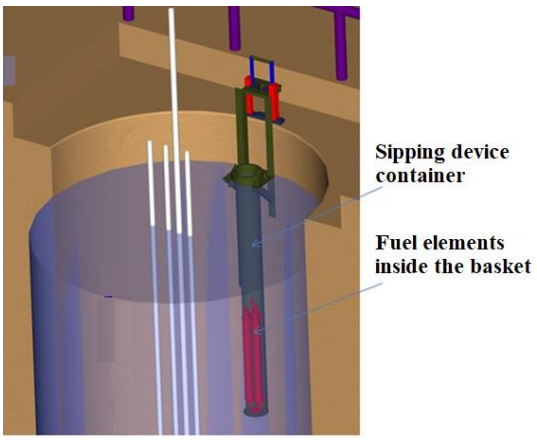

Fig. 11 - Container positioning at the pool edge loaded with three fuel elements submitted to the sipping test.

3.2. Model of diffusion of the Cs-137 in the water of the sipping container

For sipping tests on irradiated fuel elements stored for many years inside reactor pool, the most suitable fission product for use as failure monitor is cesium-137, due to its long half-life (30.14 years), great fission yields and high solubility in water [20] [21]. Thus, a correlation was developed to find the diameter of a hypothetical small cylindrical hole in the fuel cladding as a function of Cs-137 activity values to be measured.

The diffusion of cesium into a material medium can be described by Fick's law, which prescribes that the density of the diffusion flux of a substance is linearly proportional to its concentration gradient. In the case described here, the medium is the pool water of the Triga reactor. Then, the diffusion of a fission product (Cs-137) is described by the following partial differential equation:

$$f=-D\nabla c \quad (1)$$

where f is a vector magnitude representing the cesium activity flux density ($Bq.m^2.h^{-1}$), D is the diffusion coefficient and ∇c represents the concentration gradient of cesium activity in water. Therefore, the diffusion coefficient D can be defined from the Fick equation and expressed as:

$$D = f / \nabla c \quad (2)$$

The diffusion of a radioactive fission product through a given medium is described by means of the following partial differential equation [22]:

$$D.\nabla^2\rho - \lambda.\rho + S = \frac{\partial \rho}{\partial t} \quad (3)$$

where: ρ is the concentration of atoms or ions of the fission product in the medium (atoms/cm ^{3}), D is the diffusion coefficient of the fission product in the medium (cm^2/s), λ is the radioactive decay constant of the fission product (1/s), and S is the source term describing the rate of fission product generation the in the medium ($atoms/cm^3s$).

After several algebraic operations, it was found the mean radius of a hypothetical small cylindrical hole (Fig. 12) in the fuel cladding as a function of Cs-137 activity, shown in Equation 4:

$$r_0 = \frac{-1}{(\pi.(f.(\lambda.(S_0(a^2+6.D.t))))}.\frac{a.Vol.\sqrt{3}}{\sqrt{a.Vol}}\sqrt{2.\pi.f.\lambda.S_0.(a^2.+6.D).A(t)} \quad (4)$$

where:

r_0 = average radius of the equivalent cylindrical hole in the cladding;

S_0 is the initial concentration of fission product atoms in the fuel element (atoms/cm^3), measured at the moment the fault reaches the fuel , ie, when t = 0.

Vol = volume of water contained in the container of the sipping apparatus;

A(t) = activity at time *t*;

f = fraction of Cs-137 concentration present in the irradiated fuel which is limited by the leaching water in the sipping container;

λ = decay constant of Cs-137;

a = total hole length (depth);

D = diffusion coefficient of Cs-137 in the leaching water;

t = test time of the sipping test.

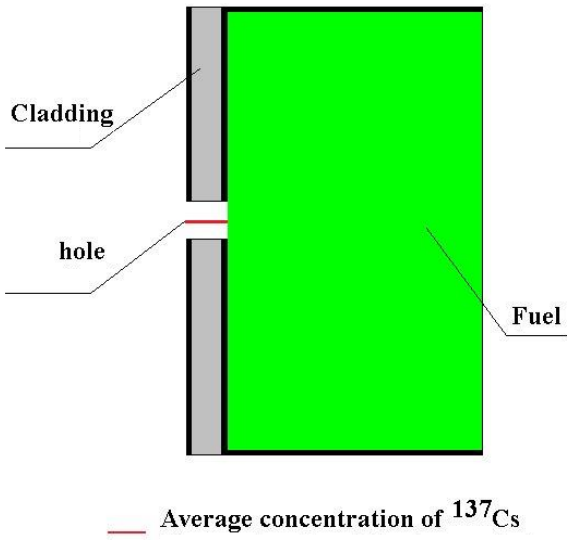

Fig. 12 - Schematic drawing showing the behavior of the Cs-137 concentration and the hypothesis of a mean concentration upon reaching equilibrium after a long time.

4. CONCLUSIONS

Sipping test is a non-destructive technique employed to evaluate the cladding structural integrity of irradiated nuclear fuels, which is based on the detection of radioactive fission products leakage to the reactor coolant, by means of gamma-ray spectroscopy. A sipping system was developed to assess the integrity of spent fuel elements irradiated in a Triga reactor of Brazil. It is necessary to keep the fission products inside the fuel elements and to prevent their escape into the environment. In the IPR-R1 Triga research reactor the fuel elements is in operation for decades and corrosion may occur causing damage of the spent fuel.

The IPR-R1 was one of the first reactors Triga to start operation in the world. It has been widely used in the training of reactor operators with practical classes with manipulation of the fuel elements. Handling causes cladding friction and can lead to damages. In visual inspection carried out in 2006 it was verified the presence of pits in the cladding of some fuel elements. With the inclusion in the core of some stainless steel-clad fuel elements together with aluminum–clad fuel elements increases the possibility of pitting corrosion due the galvanic pair formed.

In the IPR-R1 Triga reactor, despite its almost 60 years of operation, a sipping test was never performed. So it was constructed a system for performing this test according to the methodology described here. The system is ready to perform the test. A correlation was also developed between the activity of a possible release of cesium-137 in water and the diameter of a hypothetical hole. The result of this test will give subsidies for decision making regarding the need to recover or replace the failed fuel element.

Brazil has four research reactors in operation: the MB-01, a 0.1 kW critical facility; the IEA-R1, a 5 MW pool type reactor; the Argonauta, a 500 W Argonaut type reactor and the IPR-R1, a 250 kW Triga Mark I type reactor. Three of these reactors have been operating for more than

46 years, which means that they have exceeded the 40-year life expectancy range for research reactors. The methodology described here, with some adaptations, can be used to test the fuel elements of these reactors. In the future it may also be used in the Brazilian Multipurpose Reactor (RMB), in the design phase. The incorporation of procedures for the routine inspection of the fuels will increase the useful life, with reliability and safety in the operations.

ACKNOWLEDGMENTS

The authors thank the following Brazilian institutions: Nuclear Technology Development Centre (CDTN), Brazilian Nuclear Energy Commission (Cnen), Research Support Foundation of the State of Minas Gerais (Fapemig), Brazilian Council for Scientific and Technological Development (CNPq), and Coordination for the Improvement of Higher Education Personnel (Capes).

REFERENCES

[1] G.A. - General Atomics. TRIGA. In: TRIGA® Nuclear Reactors. Available from: http://www.ga-esi.com/triga/. Accessed June 27, 2017.

[2] Mesquita, A.Z.; Palma, D.A.P.; Lombardi, A.; Pereira, C.; Veloso, M.A.; Reis, P.A.L. 'Experimental investigation of thermal hydraulics in the IPR-R1 Triga nuclear reactor'. In: Amir Z. Mesquita; Iva Simcic. (Org.). Nuclear Reactors. 1ed. v. 1, pp. 1-24. DOI: 10.5772/25869. 2012.

[3] Marcum, W.R. *Thermal Hydraulic Analysis of the Oregon State TRIGA® Reactor Using RELAP5-3D*. MSc Dissertation. Oregon State University. Oregon. 2008.

[4] Simnad, M.T. 'The UZrHx alloy: its properties and use in Triga fuel'. *Nuclear Engineering and Design*, Volume 64, Issue 3, Pages 403-422. April. DOI: 10.1016/0029-5493(81)90135-7. 1981.

[5] CDTN/Cnen - Centro de Desenvolvimento da Tecnologia Nuclear, Comissão Nacional de Energia Nuclear. *Reactor Safety Analysis Report - IPR-R1 Triga Reactor*. Belo Horizonte. Brazil. 2008. (in Portuguese).

[6] ASME - The American Society of Mechanical Engineers. *ASME boiler and pressure vessel code - Materials - ASME Section II Part D - Properties (Customary) Materials*. New York, NY. 2010.

[7] IAEA - International Atomic Energy Agency. Corrosion of Research Reactor Aluminium Clad Spent Fuel in Water, Technical Reports Series No. 418, Vienna. 2003.

[8] Park, J.Y.; Shim, M.S.; Lee, J.H. 'Current Status of Integrity Assessment by Sipping System of Spent Fuel Bundles Irradiated in Candu Reactor'. *Nuclear Engineering and Technology*, Vol.46 No.6, pp 875-882. December. 2014. DOI: 10.5516/NET.09.2014.018.

[9] Mesquita, A.Z. *Experimental Investigation on Temperatures Distributions in a Research Nuclear Reactor TRIGA IPR-R1*. Ph.D. Thesis, Universidade Estadual de Campinas, São Paulo, Brazil. (in Portuguese). 2005.

[10] Alencar, D.A.; Silva Jr, S.F.; Mesquita, A.Z.; Câmara, L.O.; Oliveira, P. 'Using visual inspection NDT to check integrity of Triga mark I fuel rods'. *Proceedings of the 3nd. World Triga Users Conference*. Belo Horizonte. 2006

[11] GentiL, V. *Corrosão*. 6a ed. Rio de Janeiro. LTC - Livros Técnicos e Científicos Editora S.A. 2011.

[12] IAEA - International Atomic Energy Agency. *Operating Experience from events reported to the IAEA incident reporting system for research reactors.* Iaea-Tecdoc-1762. Vienna. 2015.

[13] Reis, P.A.L.; Costa, A.L.; Pereira, C.; Veloso, M.A.F.; Mesquita, A.Z.

Simulation of a Triga Reactor Core Blockage Using RELAP5 Code. *Science and Technology of Nuclear Installations.* Volume 2015. DOI: 10.1155/2015/354163.

[14] IAEA - International Atomic Energy Agency. *Safety Analysis For Research Reactors.* Safety Reports Series No. 55. Vienna. 2008.

[15] Sabino, C.V. (1999). *Chemical control of the cooling water of the IPR-R1 Triga reactor*. CDTN/Cnen Technical Note. Belo Horizonte. Brazil. (in Portuguese). 1999.

[16] Calazans, P.N. *Production of coconut aroma by Trichoderma harzianum using sugarcane bagasse*. Masters Dissertation. Chemical Engineering. Universidade Federal do Sergipe - UFS. Brazil. 2012. (in Portuguese)

[17] Freitas, M.M. General and morphological aspects of *trichoderma sp*.. Available at: http://fitopatologia1.blogspot.com.br/2010/12/descricao-micologicaaspectos-gerais-e.html. Access in 25 Jan 25, 2017. (in Portuguese).

[18] Rodrigues, R.R.; Mesquita, A.Z. 'Design of a System for Fuel Elements Cladding Inspection of a Nuclear Research Reactor'. *Proceedings of the 4th International Conference on Integrity, Reliability and Failure* (IRF'2013). Pp. 69-70. Funchal, Ilha da Madeira. 2013.

[19] CDTN/Cnen - Centro de Desenvolvimento da Tecnologia Nuclear, Comissão Nacional de Energia Nuclear. *Articulated crimper for handling devices and equipment at a distance*. Patent INPI Nr.: PI 0803376-5 A2. Classification: B25B 9/04. Published: in Oct 2011.

[20] Perrotta, J.A.; Terremoto; L.A.A.; Zeituni, C.A. 'Experience on wet storage spent fuel sipping at IEA-R1. Brazilian research reactor'. *Annals of Nuclear Energy* 25 (4 −5), pp. 237−258. 1998. DOI: 10.1016/S0306-4549(97)00039-X.

[21] Terremoto, L.A.A.; Seerban, R.S.; Zeituni, C.A.; Silva, J.E.R.; Silva, A.T.; Castanheira, M.; A.T.; S.M., Lucki G., Damy M.A., Teodoro C.A. 'A model for release of fission products from a breached fuel plate under wet storage'. *Progress in Nuclear Energy*. Volume 50, Issue 7, p818-827. September. 2008. DOI:10.1016/j.pnucene.2007.08.003.

[22] Friskney, C.A. and Speight, M.V. 'A calculation on the in-pile diffusional release of fission products forming a general decay chain', *Journal of Nuclear Materials.* Volume 62, Issue 1, pp89 – 94. 1976. DOI:10.1016/0022-3115(76)90286-