DEVELOPMENT OF AN IRRADIATION SYSTEM FOR RADIOISOTOPE PRODUCTION APPLIED TO INDUSTRIAL PROCESS TOMOGRAPHY

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ABSTRACT

Among the various applications of radioisotopes, the use of radiotracers is considered the most important in diagnosing operation and troubleshooting of industrial process plants in chemical and petrochemical companies. The radiotracers are used in analytical procedures to obtain qualitative and quantitative data systems, in physical and physicochemical studies transfers. In the production of gaseous radioisotopes used as tracers in industrial process measurements, argon-41 (^{41}Ar) and krypton-79 (^{79}Kr) have low reactivity with other chemical elements. ^{41}Ar is a transmitter range with high-energy (1.29MeV) and a high percentage of this energy transformation (99.1%), resulting in relatively small quantities required in relation to the other, for an efficient detection, even in large thicknesses components. Nowadays, the production of gaseous radioisotopes in nuclear research reactors is performed in small quantities (batches), through quartz ampoules containing natural gas ^{40}Ar or ^{78}Kr. In this sense, the aim of this study is to develop an irradiation system for gaseous radioisotope production in continuous scale, applied in industrial applications of emission tomography and flow measurement. The irradiation system may produce ^{41}Ar with activity of $7.4x10^{11}$Bq (20Ci) per irradiation cycle, through the Reactor IEA-R1 with 4.5MW and average thermal neutron flux of $4.71x10^{13}$ $ncm^{-2}s^{-1}$ to meet an existing demand in NDT and inspections companies, and even needed by the Radiation Technology Centre, at IPEN/CNEN-SP. The irradiation system consists of an aluminium irradiation capsule, transfer lines, needle valves, stripy connections, quick connectors, manometer, vacuum system, dewar, lead shielding, storage and transport cylinders, among other components. The irradiation system was approved in the leakage and stability tests (bubble test, pressurization, evacuation and with leak detector equipment SPECTRON 600 T). In the experimental production, alanine dosimeters were distributed into various components of the irradiation system, obtaining $1.07x10^{11}$Bq (2.9Ci) of ^{41}Ar. In addition, exposure rates were determined in the lead shielding wall, in which the liquefied radioactive gas was concentrated, and in the storage and transport cylinders after ^{41}Ar was transferred, by the portable radiation meter Teletector ® Probe 6150 AD-t/H.

Keywords : Industrial process measurement, emission tomography, radioisotope production, argon-41, irradiation system.

1 INTRODUCTION

Among the various industrial applications of radioisotopes, such as the use of fixed or incorporated sources of ionizing radiation on gammagraphy, level gauges for liquids and density or thickness measurements, the use of radionuclides as tracers (radiotracers) is considered one of the most important applications. A tracer consists of an injected material in a system to determine the actual conditions of the system in relation to the passage and location of fluid, by detecting this material at different points. There are also non isotopic chemical tracers. Fluorescent dyes and radiotracers are widely used for petrochemical industries, possessing properties associated with the emission of radiation that facilitate the analysis of complex systems, difficult to access, common in such industries (Bradford, 1953; IAEA,1990; IAEA, 2009).

Argon-41 (^{41}Ar) and kripton-79 (^{79}Kr) stand out in the production of gaseous radioisotopes used as industrial process tracers, having very low reactivity with other elements. The ^{41}Ar is a high energy gamma source (1.2MeV), with half-life of 110 minutes and high yield in the highest energy photon. These properties allow the use of relatively small portions, compared to other radioisotopes, to detect its presence in high width walls (5 to 6cm) setups. On the other hand, ^{79}Kr, with lower energy gamma (0.51MeV) but with a half-life of 35 hours, is affordable to be used in locals that are far from the isotope production center (IAEA, 2009; Charlton, 1986; Okuno and Yoshimura, 2010).

1.1 Background

The production of small quantities of gaseous radioisotopes in nuclear research reactors is carried out by quartz ampoules. Fig. 1 shows the sequential steps of India's first production neutron activation, in 2006, of ^{41}Ar and ^{79}Kr, intending to normalize the production and the delivery of radioisotopes for industrial use (Yelgaonkar et al, 2007; Quang et al, 2007).

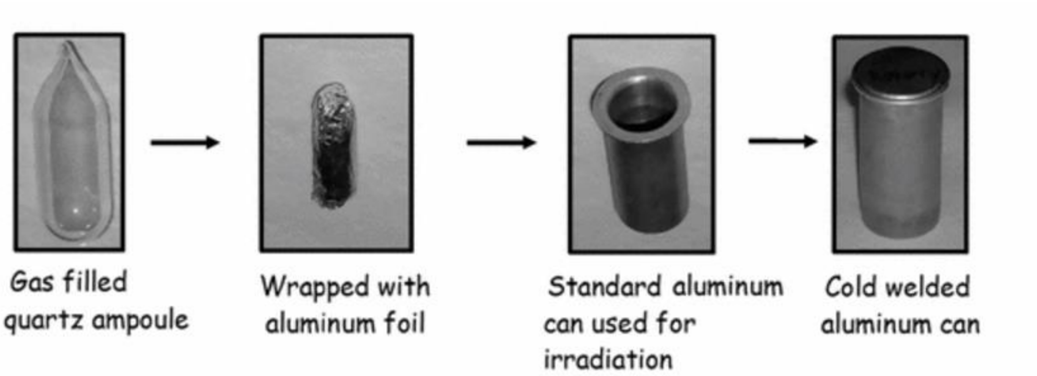

Figure 1. Gaseous radioisotope production in quartz ampoules (Yelgaonkar et al, 2007).

For higher quantity productions, irradiation setups consisting of sample cylinders, valves and transfer lines to lead shielded storage cylinders, which are taken to the radiotracer use site after the radioactive gas loading.

This second way of production was adopted for few times in the IEA-R1 nuclear research reactor, located in IPEN-CNEN/SP, by argon-40 (^{40}Ar) irradiation, transfer and transport. The setup was made in the reactor's water pool room always when the operation was done to produce the ^{41}Ar radiotracer to attend the enterprises increasing demand of non destructive tests and inspections. Due to leakage issues and high exposition to the operators, this type of production was interrupted. Thus, the development of the gaseous radioisotope production irradiation system originated the topic of this work.

1.2 Objectives

a) To develop an irradiation system with the capacity of a permanent production of gaseous radioisotopes to be used as radiotracers, in batches up to 7.4×10^{11}Bq (20Ci) using a water pool type nuclear research reactor (Material Testing Reactor - MTR), like the IEA-R1 nuclear reactor, to meet the demand for NDT and inspections for enterprises and the Radioisotope Technology Application group of the Radiation Technology Centre (CTR), at IPEN-CNEN/SP;

b) To produce ^{41}Ar or ^{79}Kr with a lower activity, to evaluate the radioprotection and production of the developed system; and

c) To allow the gaseous radiotracer production system for partner enterprises of NDT and to follow the equipment use and its performance in chemical and petrochemical industries.

2 MATERIAL AND METHODS

2.1 System components

The following material and equipment are used to develop and construct the irradiation system at the CTR and Research Reactor Centre (CRPq), both in IPEN-CNEN/SP:

a) Aluminum irradiation capsule, with 150cm³ internal volume to be positioned in the IEA-R1 nuclear reactor core, connected to a long tube. The rigid tube allows the capsule handling, inserting and removing it from the reactor core. The process of loading and unloading the gas to be irradiated to produce the gaseous radiotracer, shown in Fig. 2;

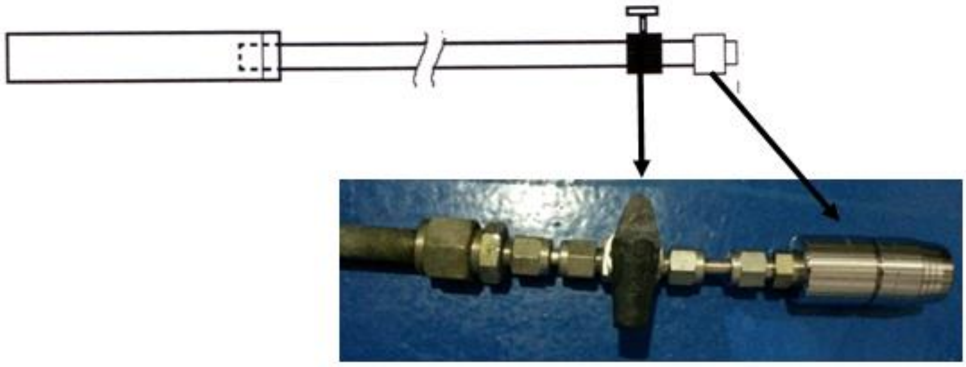

Figure 2 – Irradiation capsule with long tube (schematic) and gas load/unload control valve (in detail).

b) Solid rubber wheel platform type car;

c) Argon gas cylinder 5.0 (99,999%);

d) AISI 304 stainless steel transfer line with 6.35mm (1/4”) diameter on the superior level (Fig. 3) and 3.175mm (1/8”) on the inferior level (Fig. 4);

Figure 3. AISI 304 stainless steel tubing with 6.35mm (1/4”) inner diameter (superior level) and needle valves (V2-V8).

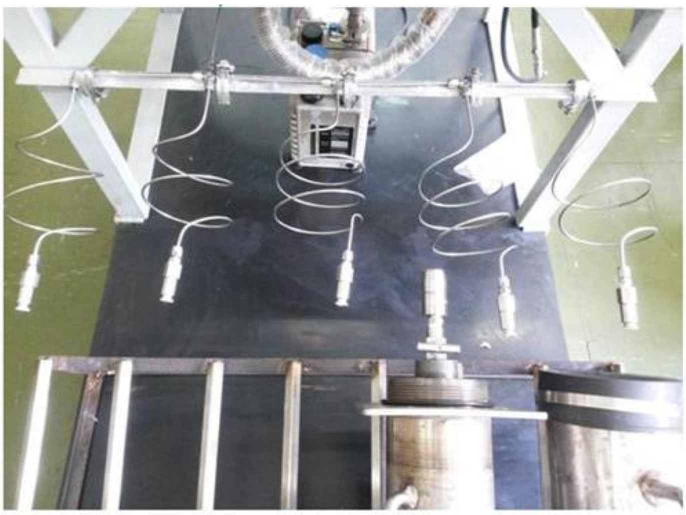

Figure 4. AISI 304 stainless steel tubing with 3.175mm (1/8”) inner diameter (inferior level).

e) Needle valves, ringed connection, fast lock connectors, manovacuometer and AISI 304 stainless steel tubing for the radioactive gas transfer line setup (Fig. 5);

Figure 5. Needle valves, ringed connection, fast lock connectors, manovacuometer and AISI 304 stainless steel tubing for the radioactive gas transfer line setup.

f) Liquid nitrogen based freezing system (dewar*)* for gas liquefaction (Fig. 6);

Figure 6. Liquid nitrogen based freezing system for gas liquefaction.

g) Dewar with 11cm thick walls lead shielding for the liquefaction (Fig. 7) (National Institute of Standards and Technology, 2016);

Figure 7. Lead shielding for the liquefaction dewar.

h) Acrylic box to isolate the transfer lines with air exit to be connected with the IEA-R1 nuclear reactor exhaust system (Fig. 8);

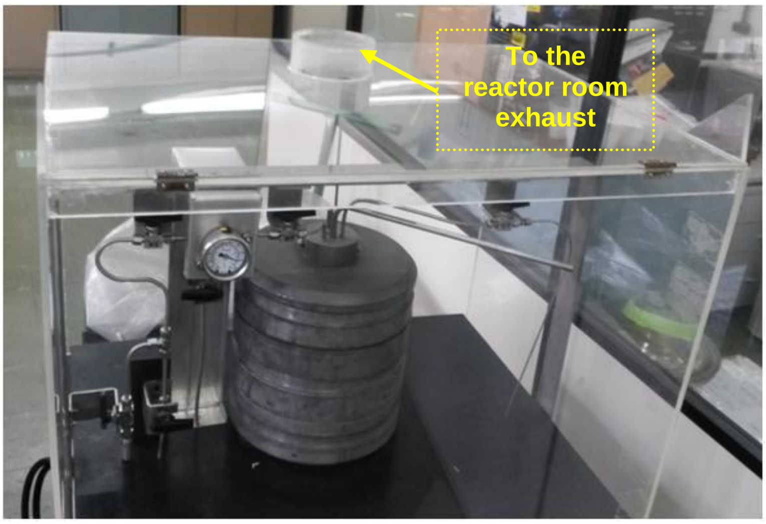

Figure 8. Acrylic box with inner air exit to the IEA-R1 nuclear reactor exhaust system.

i) Vacuum system with the mechanical vacuum pump; and

j) Cylinder for storage and transport of the gaseous radiotracers (^{41}Ar or ^{79}Kr) in AISI 304 stainless steel, with 5cm thick lead jackets coupled to needle valves and fast lock connections on both sides, and with 20cm³ of inner volume (Fig. 9).

Figure 9. Cylinder for storage and transport of the gaseous radiotracers.

2.2 Methodology

The irradiation system was designed and mounted in the platform type cargo car.

To assure the tightness of the system against radioactive gas leakage, and improve the overall radioprotection, three leak test proceedings were performed:

- **a)** Foam former solution bubble testing;
- **b)** Pressure tracking on the pressurized and evacuated system; and
- **c)** Helium leak detection procedure.

For the gaseous radioisotope production, the neutron activation technique was used, placing the irradiation ampoule inside the IEA-R1 reactor water pool (Fig. 10). The ampoule is placed on a known thermal neutron flux. The nuclear reaction described in Equation 1 occurs with a neutron capture and a prompt gamma is released, then, a radioisotope is formed (Kaplan, 1966).

$$_{Z}X^{A} + _{0}n^{1} \longrightarrow [_{Z}X^{A+1}] \longrightarrow (_{Z}X^{A+1})^{*} + \gamma_{prompt} \qquad (1)$$

Figure 10. IEA-R1 nuclear reactor water pool and the ampoule inserted.

2.3 Experimental sequence

The development of the gaseous radioisotope irradiation system used the following steps:

- **a)** Irradiation system design with gas transport system dimensioning, calculations of the necessary radiation shielding and system parts specifications;
- **b)** System parts production and device mounting on the platform type cargo car;
- **c)** Operational proceedings determination;
- **d)** Performance, stability and tightness tests;
- **e)** Irradiation system technical and operational specifications is submitted to the Internal Safety Committee of the IEA-R1 nuclear reactor; and
- **f)** Thermal Neutron flux exposure to get a 1.0×10^{11}Bq (2.9Ci) of ^{40}Ar gas to check the system performance before starting the production of batches with up to 740GBq (20Ci) in industrial scale.

3 RESULTS

3.1 Irradiation system design and construction

The irradiation system was designed to allow the loading, neutron flux exposition, unloading of the gas on an irradiation capsule (IC) located near the nuclear reactor core and transferring it to storage and transport cylinders (STC). It uses the liquid nitrogen frozen gas liquefaction, vaporization with heating to normal room temperature and valves dealing to conduct the gas to the STCs.

Considering movement in the IEA-R1 reactor's water pool room, most of the system is mounted in the cargo car with the parts described. There is, also, an acrylic box intended to isolate eventual gas leaks from the room and take the air inside the box to the reactor's exhaust system.

Fig. 11 shows the conception design of the outside pool part, already coupled to the STCs and the argon gas cylinder 5.0 (99,999%).

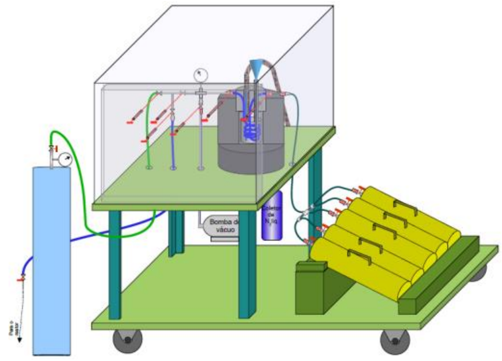

Figure 11 – Industrial processes applied to gaseous radioisotope production irradiation system.

Since the material and parts acquisition, the acrylic box mounting and the manufacturing of the lead shielding, the system was mounted in the cargo car. The complete system is shown in Fig. 12.

Figure 12 – System to load and unload the irradiation capsule placed inside the IEA-R1's water pool.

3.2 Operational routine

The ^{41}Ar producing operational proceeding was established and it was divided in four steps, as seen in Fig. 13. The proceedings took place after all the connections had been made, including the rigid long tube to place the irradiation ampoule and the STCs, and the exhaust duct was connected on the top of the acrylic box.

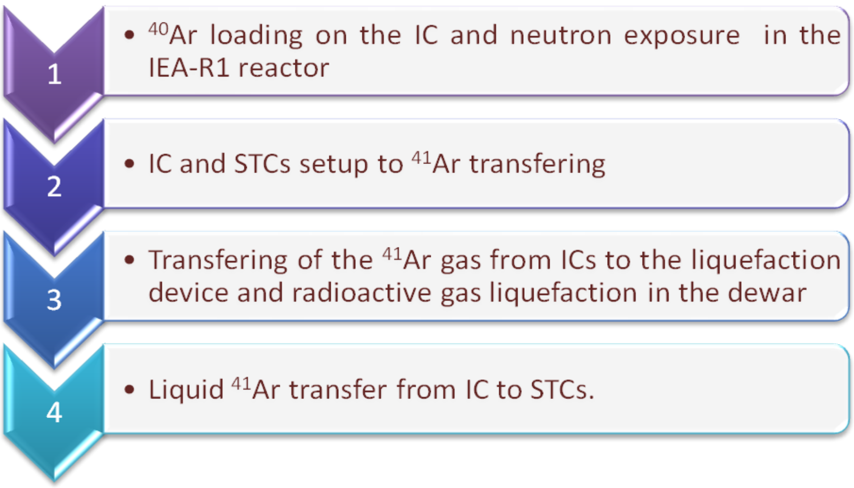

Figure 13 - ^{41}Ar production proceeding steps.

3.3 Testing tightness and stability FROM HERE

The three techniques of leak detection described in 2.2 session were performed and it was concluded that there is no leak in the irradiation system. The tests showed that the detected leak rates ranged from $1x10^{-9}$ to $5x10^{-9}$ mbar.L/s (10^{-4} µPa.m³.s^{-1}).

3.4 Irradiation for system testing

An irradiation test for the system was performed in the ^{40}Ar to obtain ^{41}Ar by neutron irradiation. The iradiation capsule was positioned in a $4.71x10^{13}$ $ncm^{-2}s^{-1}$ average thermal neutron flux site inside the IEA-R1 nuclear reactor. This exposure intended to get about $1.0x10^{11}$Bq (2.9Ci) of ^{41}Ar activity.

The irradiation data are shown in Table 1.

Table 1 - ^{41}Ar testing production with the irradiation system.

Isotope	^{40}Ar
Pressure inside the ampoule	1.5bar
Estimated mass	0.37g
Irradiation time	4h
Decay time	0.6h (36min)
Irradiation site	34A
Mean thermal flux	$4.71x10^{13}$ $ncm^{-2}s^{-1}$
Estimated activity	$1.07x10^{11}$Bq (2.9Ci)

The ^{41}Ar produced in this test was stored in only one STC. The irradiation system was observed, 10 alanine dosimeters were used to evaluate the radiation exposure through the system and the dose rate outside the shielding was measured when the liquefied radioactive gas was concentrated in it.

The alanine dosimeters were evaluated with the ^{41}Ar (1.29MeV) energy calibration curve. Nine dosimeters showed that the doses were below detection limit (5Gy). The tenth one, located between the STCs transfer tubes (Fig. 14) showed a 5Gy absorbed dose.

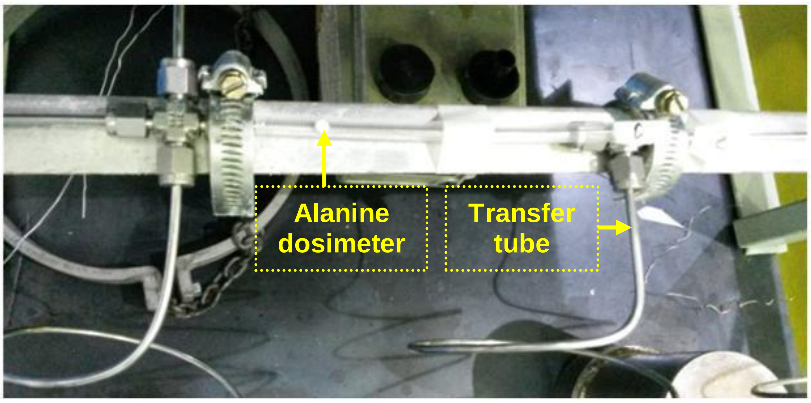

Figure 14 – Alanine dosimeter positioned between the transfer tubes.

The measured dose rates were 10mSv/h on the shielding surface when the radioactive liquefied gas was concentrated in it and 25mSv/h on the STC surface, after 41Ar transfer to it.

4 DISCUSSION AND CONCLUSIONS

To develop the irradiation system according to the design, the first objective was successfully achieved.

Related to the operational proceedings, tightness and stability of the irradiation system, the conclusions are:

- The irradiation system is ready to be used according to the proceedings, since the tightness and stability tests showed that no component presented performance issues;
- The system is tight because the leak detector equipment showed 10^{-4} µPa.m³.s^{-1}, which is 100 times lower than the determined limit in the ISO 9978 – *Sealed radioactive sources – Leakage test methods* (1992); and
- The system tightness was, also, proved because no radioactive gas was detected by the sensors in the IEA-R1 nuclear reactor exhaust system, during the test performance.

Performing the test irradiation to obtain the ^{41}Ar with the irradiation system, the assumptions are:

- The alanine dosimeter placed between the STCs transfer lines showed the highest absorbed dose (5Gy). The lines inner volume, although very small, may retain some radioactive gas;
- The highest exposure rates obtained in the dewar shielding (10mSv/h) and STC surface (25mSv/h). To limit the operators dose, the valves handling should be very fast, keeping distance and/or replacing the valves with electromagnetic ones; and

The irradiation system developed produces gaseous radioisotopes (^{40}Ar and ^{78}Kr) in continuous scale, which can be applied in industrial applications of emission tomography and flow measurement (Johansen and Jackson, 2004).

REFERENCES

BRADFORD, J.R., (1953), *Radioisotopes in industry.* Reinhold; Chapman & Hall: New York, 1953.

CHARLTON J.S., (1986), *Radioisotope techniques for problem-solving in industrial process plants, Le onard Hill.* ISBN-13: 978-94-010-8306-5.

INTERNATIONAL ATOMIC ENERGY AGENCY, (1990), *Guidebook on Radioisotope Tracers in Indust ry, Technical Reports Series, 316*. ISBN 92-0-165090-6.

INTERNATIONAL ATOMIC ENERGY AGENCY, (2009), *Leak Detection in Heat Exchangers and Underground Pipelines Using Radiotracers.*

INTERNATIONAL ORGANIZATION FOR STANDARDIZATION, (1992), *Radiological protection – Sealed radioactive sources – Leak test methods.* ISO 9978-1992.

JOHANSEN G.A.; JACKSON P., (2004), *Radioisotope Gauges for Industrial Process Measurements,* John Wiley & Sons, Ltd. ISBN 0-471-48999-9.

KAPLAN, I., (1963), *Nuclear Physics*. 2. ed. Massachusetts: Addison-Wesley Publishing Company Inc.

NATIONAL INSTITUTE OF STANDARDS AND TECHNOLOGY, (2016), *X-Ray Mass Attenuation Coefficients.* Available in: http://physics.nist.gov/PhysRefData/XrayMassCoef/chap2.html. Access in: 03 Feb. 2016.

OKUNO, E.; YOSHIMURA, E.M., (2010), *Radiation Physics*. 1 ed. Sao Paulo: Oficina de textos.

QUANG, N.H.; TRI, B.Q.; NGAN, H.T.K.; DUY, D.N.; HAI, T.T.; HA, P.H.C.; COUNG, T.B.; KHOA, P.T., (2007), *Establishment of Tracer Technologies in Gas Phase,* In: The Annual Report. Nuclear Research Institute, VAEC, Vietnam.

V.N. YELGAONKAR; K.C. JAGADEESAN; V. SHIVARUDRAPPA; V.K. SHARMA; S. CHITRA, (2007), Production of ^{41}Ar and ^{79}Kr gaseous radiotracers for industrial applications. *Journal of Radiationalytical and Nuclear Chemistry*, 274, 2, pp. 277-280.