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Irradiation system for production of gaseous radioisotopes used as tracers in industrial process measurements

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ABSTRACT

The use of radioisotopes as radiotracers is considered the most important in diagnosing operation and troubleshooting of industrial process plants in chemical and petrochemical companies. They 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) stand out because each has low reactivity with other chemical elements. ^{41}Ar is a transmitter range with high-energy (1.29 MeV) 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 7.4×10^{11} Bq (20 Ci) per irradiation cycle, through the Reactor IEA-R1 with 4.5 MW and average thermal neutron flux of 4.71×10^{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, ringed 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 obtaining 1.07×10^{11} Bq (2.9 Ci) of ^{41}Ar, alanine dosimeters were distributed into various components of the irradiation system. 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.

1. Introduction

Among the various industrial applications of radioisotopes, such as the use of fixed or incorporated sources of ionizing radiation on industrial radiography, 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, for example using the method of the residence time distribution (RTD), according Fig. 1, where the concentration-time curves at the input $C_i(t)$ and at the output $C_o(t)$ are recorded by means of detectors. 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; IAEA, 2001).

The choice of radiotracers depends on many factors (specific activity, half-life, type of radiation emitted, energy level) and the radioisotopes argon-41 (^{41}Ar) and kripton-79 (^{79}Kr) stand out in the production of gaseous radioisotopes used as industrial process tracers according to Table 1, having very low reactivity with other elements. The ^{41}Ar is a high energy gamma source (1.2 MeV), with half-life of 110 min and high yield in the highest energy photon. These properties allow the use of relatively small portions, compared to other

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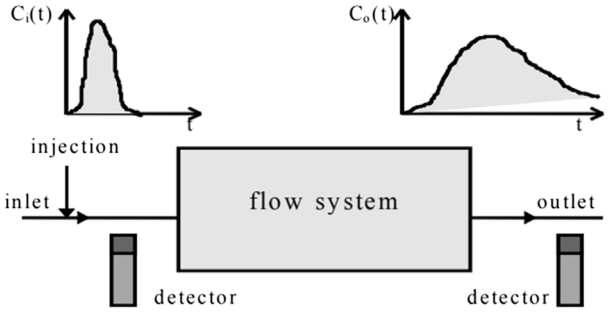

Fig. 1. RTD principle (IAEA, 2001).

Table 1
Radiotracer commonly used for leak detection in heat exchangers (IAEA, 2009).

Radioisotope	Half-life	Gamma Energy MeV (Abundance %)	Chemical Form	Tracing Phase
Sodium-24	15 h	1.37 (100%); 2.75 (100%)	Sodium carbonate	Aqueous
Bromine-82	36 h	0.55 (70%) 1.32 (27%)	Ammonium bromide, Methylbromide, Dibromobenzene	Aqueous Gases Organic
Iodine-131	8.04 d	0.36 (80%) 0.64 (9%)	Potassium or sodium iodide, Iodobenzene, Hippuran	Aqueous Organic
Technetium-99m	6 h	0.14 (90%)	Pertechnetate	Aqueous
Indium-113m	100 min	0.392 (65%)	EDTA complex	Aqueous
Krypton-85	10.6 y	0.51(0.7%)	Krypton	Gases
Krypton-79	35 h	0.51 (15%)	Krypton	Gases
Xenon-133	5.27 d	0.081 (37%)	Xenon	Gases
Argon-41	110 min	1.29 (99%)	Argon	Gases

radioisotopes, to detect its presence in high thickness walls (5–6 cm) setups. On the other hand, ^{79}Kr, with lower energy gamma (0.51 MeV) but with a half-life of 35 h, is affordable to be used in industrial process plants in chemical and petrochemical companies that are far from the isotope production centre (IAEA, 2009; Charlton, 1986; Okuno and Yoshimura, 2010).

The production of argon-41 and krypton-79 occurs by nuclear reactions with thermal neutrons (capture reaction) in a research reactor, such as the IEA-R1 in Nuclear and Energy Research Institute (IPEN-CNEN/SP). The nuclear reaction occurs with a neutron capture and a prompt gamma is released, then, a radioisotope is formed, according Equations (1) and (2) e 3. (Kaplan, 1963).

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

$$ {}^{40}Ar + n \rightarrow {}^{41}Ar + \gamma_{prompt} \quad (2) $$

$$ {}^{78}Kr + n \rightarrow {}^{79}Kr + \gamma_{prompt} \quad (3) $$

In addition, a estimated activity during a irradiation with neutrons can be obtained by means Equation (4), where N_1 represents the number of atoms of the stable isotope, "σ" the cross-section to reaction (n,γ), "ϕ" the thermal neutrons flux, "λ" the radioactive decay constant of the radioisotope produced and "t_i" the time of irradiation.

$$ A = N_1 \sigma \Phi \left(1 - e^{-\lambda t_i}\right) \quad (4) $$

Argon gas mixtures has three stable isotopes ^{36}Ar (0.337%), ^{38}Ar (0.063%) and ^{40}Ar (99.6%). Since the (n,) products of the first two isotopes are long-lived, the cross section of particular interest is that of the most abundant ^{40}Ar, the (n,) product being 1.83 h ^{41}Ar and gamma energy 1.29 MeV (abundance 99,1%) (Ranakumar et al., 1969).

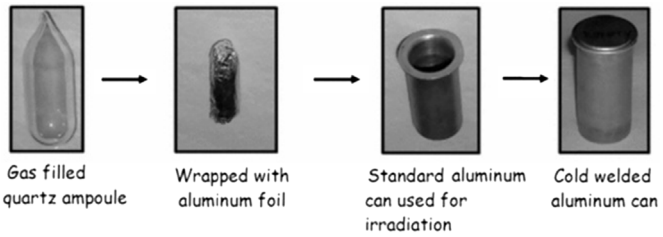

Fig. 2. Gaseous radioisotope production in quartz ampoules (Yelgaonkar et al., 2007).

1.1. Background

The production of small quantities of gaseous radioisotopes in nuclear research reactors is carried out by quartz ampoules. Fig. 2 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).

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, according to Fig. 3. Due to leakage issues and high exposure to the operators, this type of production was discontinued. 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 (20 Ci) using the water-cooled test Research Reactor IEA-R1 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 150 cm^3 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. 4;
b) Solid rubber wheel platform type car;
c) Argon gas cylinder 5.0 (99,999%);
d) AISI 304 stainless steel transfer line with 6.35 mm (1/4″) diameter

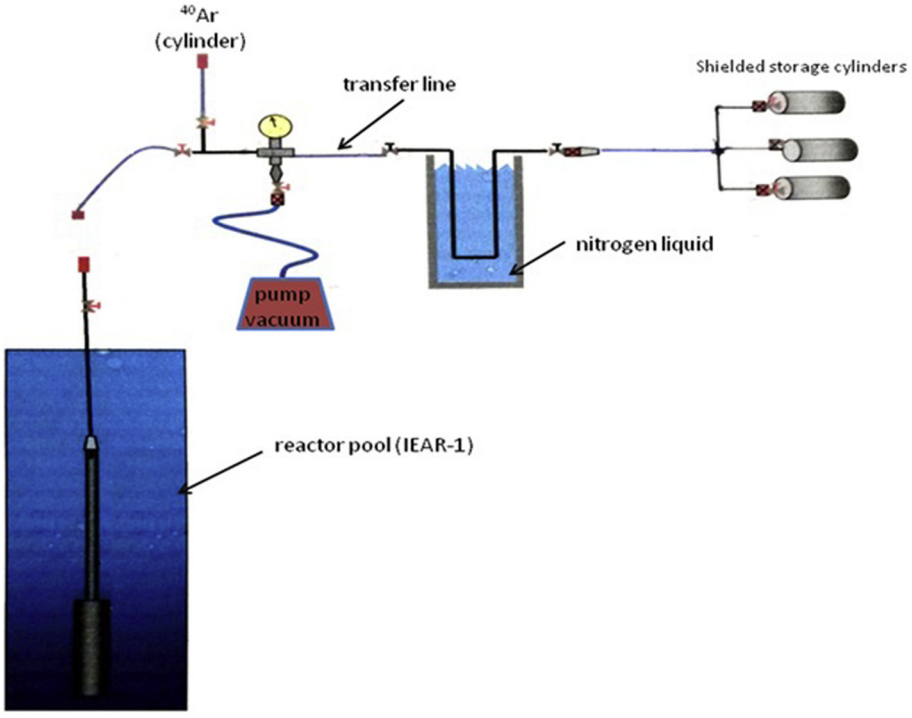

Fig. 3. Irradiation setup for ^{41}Ar production in the IEA-R1 nuclear research reactor.

on the superior level (Fig. 5) and 3.175 mm (1/8″) on the inferior level (Fig. 6);

e) Needle valves, ringed connections, fast lock connectors, manovacuometer and AISI 304 stainless steel tubing for the radioactive gas transfer line setup (Fig. 7);
f) Liquid nitrogen based freezing system (dewar) for gas liquefaction (Fig. 8);
g) Dewar with 11 cm thick walls lead shielding for the liquefaction (Fig. 9) (National Institute of Standards and Technology, 2016);
h) Acrylic box to isolate the transfer lines with air exit to be connected with the IEA-R1 nuclear reactor exhaust system (Fig. 10);
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 5 cm thick lead jackets coupled to needle valves and fast lock connections on both sides, and with 20 cm^3 of inner volume (Fig. 11).

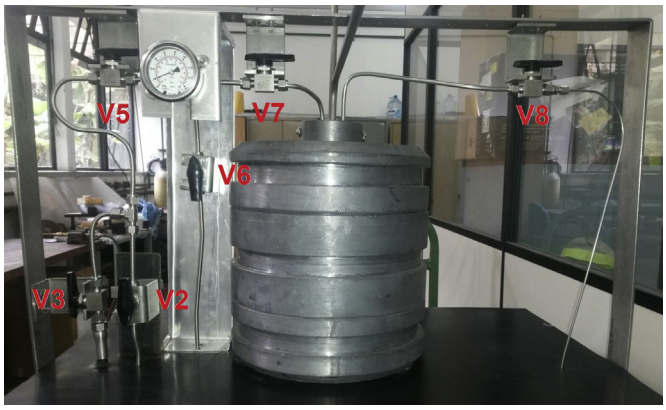

Fig. 5. AISI 304 stainless steel tubing with 6.35 mm (1/4″) inner diameter (superior level) and needle valves (V2-V8).

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.

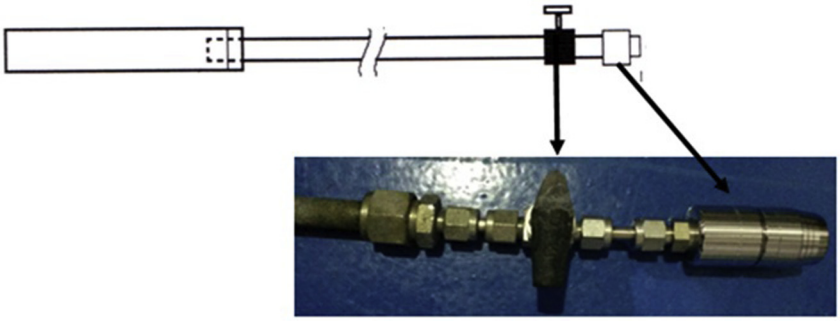

Fig. 4. Irradiation capsule with long tube (schematic) and gas load/unload control valve (in detail).

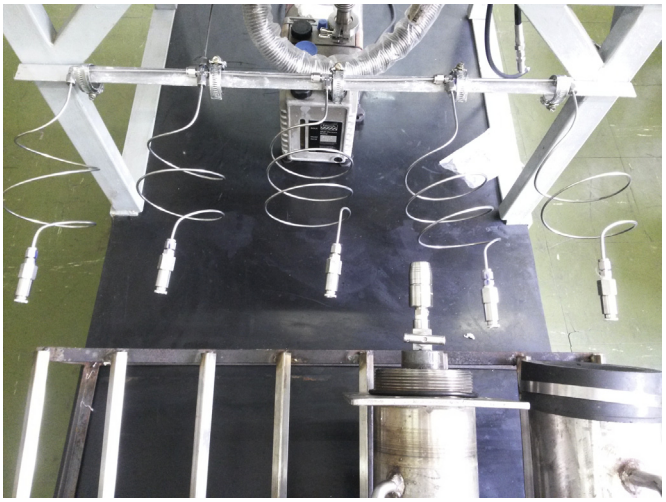

Fig. 6. AISI 304 stainless steel tubing with 3.175 mm (1/8″) inner diameter (inferior level).

For the gaseous radioisotope production, the neutron activation technique was used, placing the irradiation ampoule inside the IEA-R1 reactor water pool (Fig. 12). The ampoule is placed on a known thermal neutron flux.

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) Operationalproceedingsdetermination;
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 (2.9 Ci) of ^{40}Ar gas to check the system performance before starting the production of batches with up to 740 GBq (20 Ci) in industrial scale.

3. Results

3.1. Irradiation system design and construction

The irradiation system was designed to allow the loading, neutron flux exposure, 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.

Fig. 8. Liquid nitrogen based freezing system for gas liquefaction.

Fig. 9. Lead shielding for the liquefaction dewar.

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

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. 14.

3.2. Operational routine

The ^{41}Ar producing operational proceeding was established and it was divided in four steps, as seen in Fig. 15. The proceedings took place after all the connections had been made, including the rigid long tube to

Fig. 7. Needle valves, ringed connections, fast lock connectors, manovacuometer and AISI 304 stainless steel tubing for the radioactive gas transfer line setup.

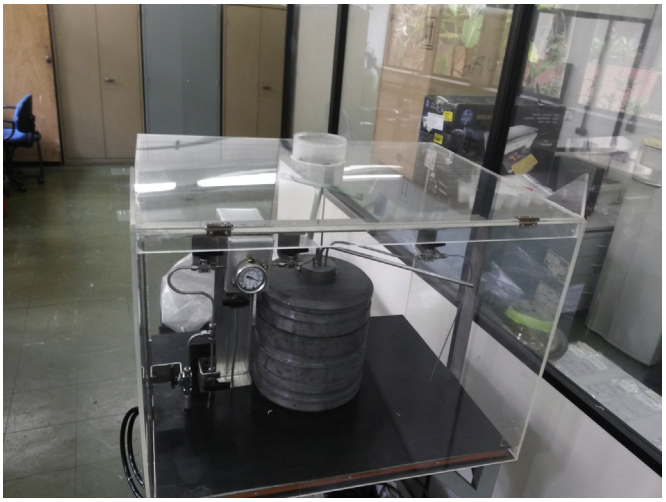

Fig. 10. Acrylic box with inner air exit to the IEA-R1 nuclear reactor exhaust system.

Fig. 11. Cylinder for storage and transport of the gaseous radiotracers.

place the irradiation ampoule and the STCs, and the exhaust duct was connected on the top of the acrylic box.

3.3. Testing tightness and stability

Three techniques of leak detection described in session 2.2 Methodology were performed. Tests showed that the detected leak rates ranged from 1×10^{-9} to 5×10^{-9} mbar.L/s (10^{-4} µPa m^3.s^{-1}). According the standard ISO 9978 - Sealed radioactive sources – Leakage test methods (1992) the system can be considered without significant leak rate. The standard establishes for sealed sources of leachable or gaseous content the limit value for the helium leakage rate of 10^{-2} µPa m^3.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 irradiation capsule was positioned in a 4.71×10^{13} ncm^{-2}s^{-1} average thermal neutron flux site inside the IEA-R1 nuclear reactor. This exposure intended to get about 1.0×10^{11} Bq (2.9 Ci) of ^{41}Ar activity. The irradiation data are shown in Table 2.

The ^{41}Ar produced in this test with estimated activity of 1.07×10^{11} Bq (2.9 Ci) was stored in only one STC, 0.6 h (36 min) after irradiation in order to wait for the decay of the short half-lives radioisotopes, such as the ^{28}Al generated with the activation of the irradiation capsule. In the ^{41}Ar experimental production, 10 alanine dosimeters were distributed into various components of the irradiation system. They were used to evaluate the dose distribution through the system. On the other hand, exposure rates were determined by a portable radiation meter Teletector® Probe 6150 AD-t/H outside the lead shielding wall, in which the liquefied radioactive gas was concentrated and in the STC wall, after ^{41}Ar was transferred.

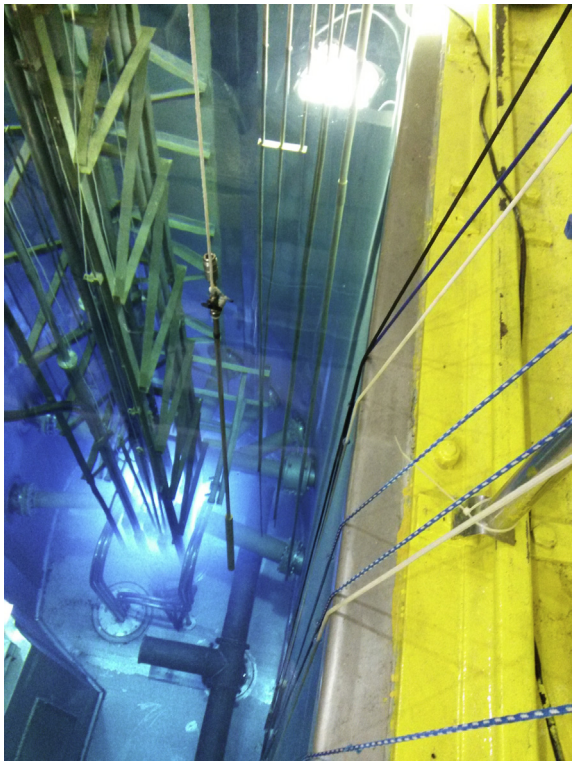

Fig. 12. IEA-R1 nuclear reactor water pool and the ampoule inserted.

The alanine dosimeters were evaluated with the ^{41}Ar (1.29 MeV) energy calibration curve. Nine dosimeters showed that the doses were below detection limit (5 Gy). The tenth one, located between the STCs transfer tubes (Fig. 16) showed a 5 Gy absorbed dose in the experimental production of ^{41}Ar, with estimated activity of 1.07×10^{11} Bq (2.9 Ci).

The measured exposure rates were 10 mGy/h on the dewar shielding surface, with almost all the radioactive gas liquefied in it, and 25 mGy/h on the STC surface, after the ^{41}Ar 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 m^3.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:

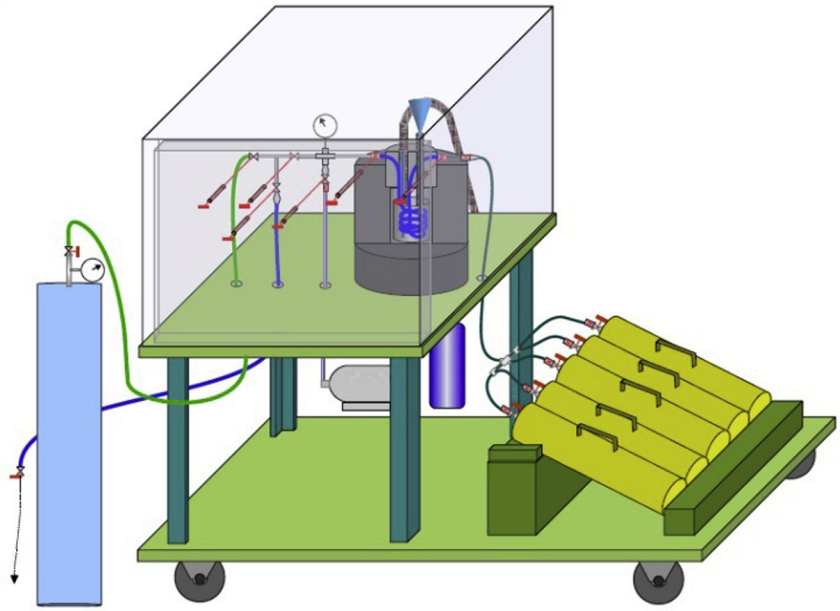

Fig. 13. Industrial processes applied to gaseous radioisotope production irradiation system.

Fig. 14. System to load and unload the irradiation capsule placed inside the IEA-R1 reactor water pool.

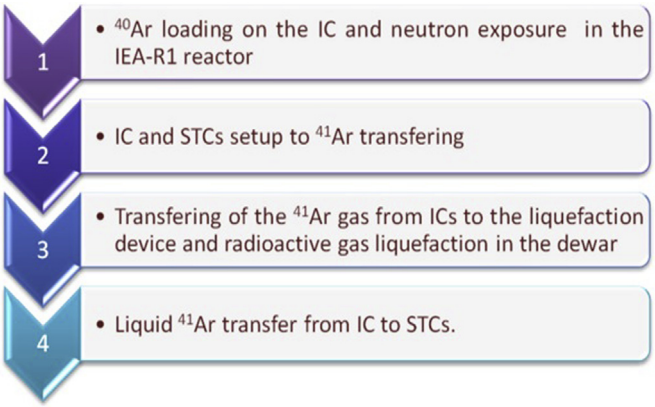

Fig. 15. ^{41}Ar production proceeding steps.

Table 2
^{41}Ar testing production with the irradiation system.

Isotope	^{40}Ar
Pressure inside the ampoule	1.5 bar
Estimated mass	0.37 g
Irradiation time	4 h
Decay time	0.6 h (36 min)
Irradiation site	34 A
Mean thermal flux	4.71×10^{13} ncm^{-2}s^{-1}
Estimated activity	1.07×10^{11} Bq (2.9 Ci)

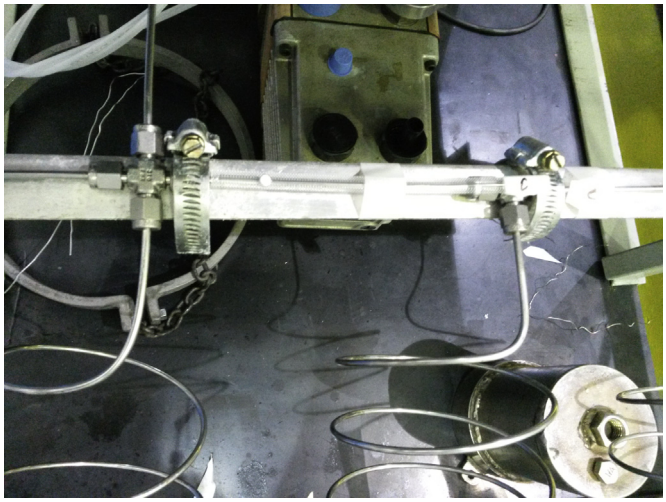

Fig. 16. Alanine dosimeter positioned between the transfer tubes.

- The alanine dosimeter placed between the STCs transfer lines showed the highest absorbed dose (5 Gy). The lines inner volume, although very small, may retain some radioactive gas;
- The highest exposure rates were obtained in the dewar shielding (10 mGy/h) and STC surface (25 mGy/h) by a portable survey meter. 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).

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