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PRELIMINARY STUDIES ON THE DEVELOPMENT OF AN AUTOMATED IRRADIATION SYSTEM FOR PRODUCTION OF GASEOUS RADIOISOTOPES APPLIED IN INDUSTRIAL PROCESSES

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ABSTRACT

The purpose of the present study is to demonstrate how it will be enhanced an Irradiation System (IS) developed with national technology to produce gaseous radioisotopes, by means of the components automation, to avoid the radiation exposure rate to operators of the system, following the ALARA principle (As Low As Reasonably Achievable). Argon-41 (^{41}Ar) and krypton-79 (^{79}Kr) can be produced in continuous scale, gaseous radioisotopes used as radiotracers in industrial process measurements and it can be used in analytical procedures to obtain qualitative and quantitative data systems or in physical and physicochemical studies transfers. The production occurs into the IS, installed in the pool hall of a nuclear research reactor in which the irradiation capsule is positioned near the reactor core containing the isotope gaseous pressurized (^{40}Ar or ^{78}Kr), by (n,γ) reaction and generate the radioisotopes. After the irradiation, the gaseous radioisotope is transferred to the system and, posteriorly, to the storage and transport cylinders, that will be used in an industrial plant. In the first experimental production, was obtained 1.07×10^{11} Bq (2.9 Ci) of ^{41}Ar distributed in two storage and transport cylinders, operating the IEA-R1 Research Reactor with 4.5 MW and average thermal neutron flux of 4.71×10^{13} n.cm^{-2}.s^{-1}. However, the system has capacity to five storage and transport cylinders and the estimated maximum activity to be obtained is 7.4×10^{11} Bq (20 Ci) per irradiation cycle. In this sense, the automation will be based in studies of the production process in the system and the use of Programmable Logic Controllers (PLC), and supervisory software allowing a remote control and consequently better security conditions.

1. INTRODUCTION

There is a wide range of industrial applications of radioisotopes, such as the use of fixed or integrated sources of ionizing radiation on industrial radiography, level gauges for liquids and density or thickness measurements, but 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 its actual conditions in relation to the passage and location of fluid, by detecting this material at different points as a function of time. There are also non isotopic chemical tracers. Fluorescent dyes and radiotracers are widely used for petrochemical industries, have properties associated with the emission of radiation that facilitate the analysis of complex systems, difficult access, common in such industries [1-3].

The Argon-41 (^{41}Ar) and kripton-79 (^{79}Kr) stand out in the production of gaseous radioisotopes used as industrial process tracers, have very low reactivity with other elements. The ^{41}Ar is a high energy gamma source (1.2 MeV), with half-life of 110 minutes and high yield in the highest energy photon. These properties allow the use of relatively small volume, compared to other radioisotopes, to detect its presence in high width steel walls (5 to 6 cm) setups. On the other hand, ^{79}Kr, with lower energy gamma (0.51 MeV) but with a half-life of 35 hours, is affordable to be used in locals that are far from the isotope production center [3-5].

1.1 Background

The production of the gaseous radioisotopes can be carried out by two techniques. The first, consists in irradiating quartz ampoules, as shown in Fig.1, that store the gaseous isotope (^{40}Ar or ^{78}Kr), in a nuclear research reactor. It has, posteriorly, to be broken in a special device, to inject the radiotracer into the industrial equipment to be analyzed, as shown in Fig.2, with the injection of ^{41}Ar or ^{79}Kr in tubes and vessels submitted to low or medium pressures up to 50 bar [6].

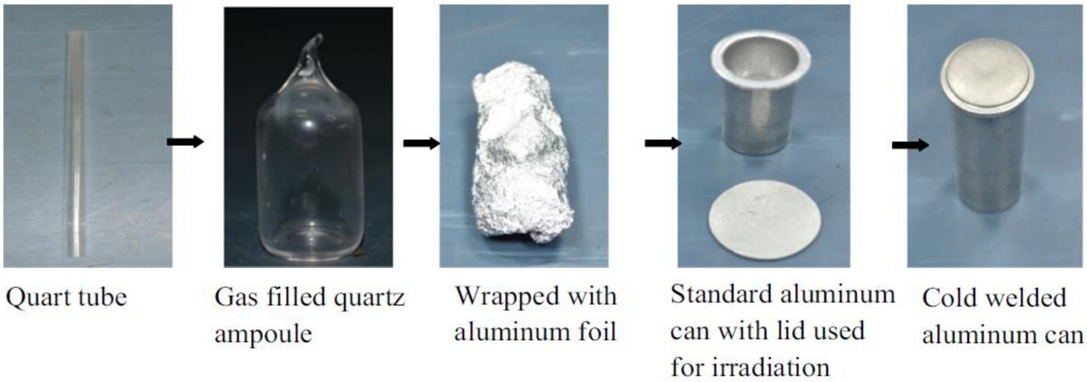

Figure 1: Stages involved in the sealing of the quartz ampoule containing noble gas in an aluminum irradiation vessel [7].

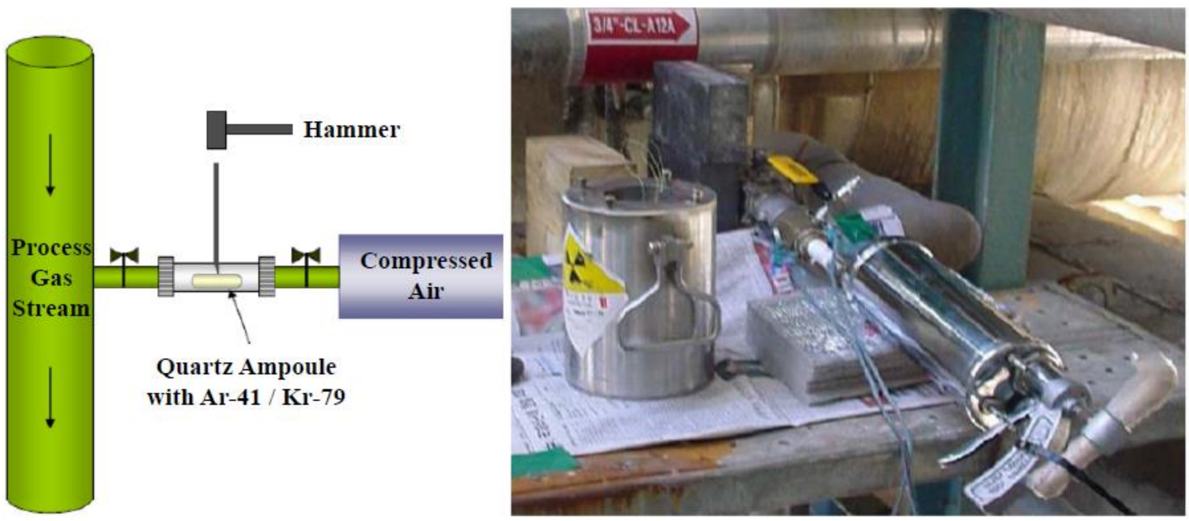

Figure 2: Injection of the gaseous radioisotopes (^{41}Ar or ^{79}Kr) in tubes and vessels submitted to low or medium pressures [6].

The second technique involves the use of irradiation capsules, as shown in Fig.3, which provide higher volumes of noble gas, but require a transfer line to other devices (lead shielded storage cylinders) that are used directly in industrial plants. Sometimes, this way of production was adopted in the IEA-R1 Nuclear Research Reactor, installed at IPEN-CNEN/SP, by argon-40 (^{40}Ar) irradiation, transfer and transport to the petrochemical company. 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 increasing demand of non destructive tests and inspections in the market. Due to leakage issues and high exposition to the operators, this type of production was interrupted. Thus, it was developed a gaseous radioisotope production irradiation system, in process of patenting (BR1020170200221), and that will be automated, according the present study [8].

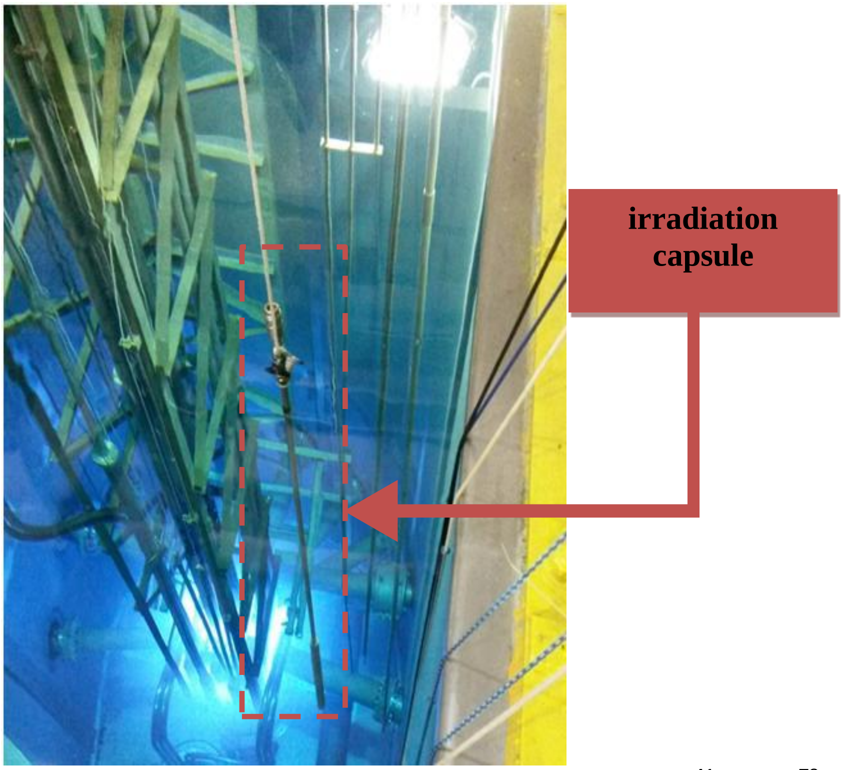

Figure 3: Irradiation capsule to produce gaseous radioisotopes (^{41}Ar or ^{79}Kr) positioned near the core of the IEA-R1 Nuclear Reactor at IPEN-CNEN/SP [8].

1.1.1 Irradiation system for production of gaseous radioisotopes - presently

The Irradiation System (IS), as shown in Fig. 4, was developed in the Radiation Technology Centre at IPEN-CNEN/SP. It may produce in continuous scale argon-41 (^{41}Ar) and krypton-79 (^{79}Kr), gaseous radioisotopes used as radiotracers in industrial process measurements, in analytical procedures to obtain qualitative and quantitative data systems or in physical and physicochemical studies transfers, because they have low reactivity with other chemical elements. Argon-41 is a gaseous radiotracer 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 steel components, while krypton-79 has a superior half-life (35 hours) than the ^{41}Ar (1,8 hours), but its emission energy is lower (510 keV) [2,3,5,9].

It 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 [9].

Figure 4: Irradiation system for production of gaseous radioisotopes [9].

Hereafter all the components of the irradiation system are listed:

a) Aluminum irradiation capsule, with 150 cm³ internal volume to be positioned near the core of the IEA-R1 Nuclear Reactor, connected to a long aluminum tube. The rigid tube allows the capsule handling, inserting and removing it from the reactor core. The irradiation capsule, long tube and control valve for loading and unloading the gas to be irradiated to produce the gaseous radiotracer are shown in Fig. 3 and 5 [9];

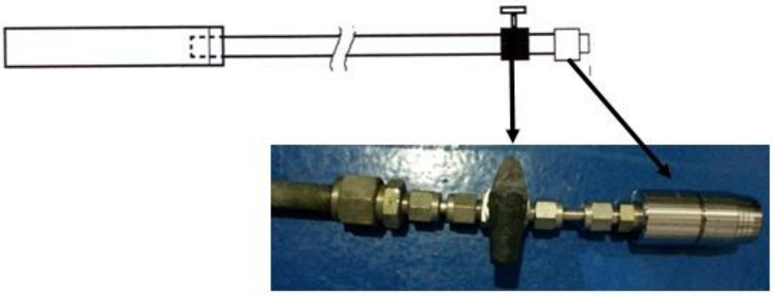

Figure 5: Irradiation capsule with long tube (schematic) and gas load/unload control valve (in detail) [9].

b) Solid rubber wheel platform type car (Fig.4) [9];

c) Argon gas cylinder 5.0 (99.999%) [9];

d) AISI 304 stainless steel transfer line with 6.35 mm (1/4") diameter on the superior level (Fig.6) and 3.175 mm (1/8") on the inferior level (Fig.7) [9];

Figure 6: AISI 304 stainless steel tubing with 6.35 mm (1/4") inner diameter (superior level) and needle valves (V2-V8) [9].

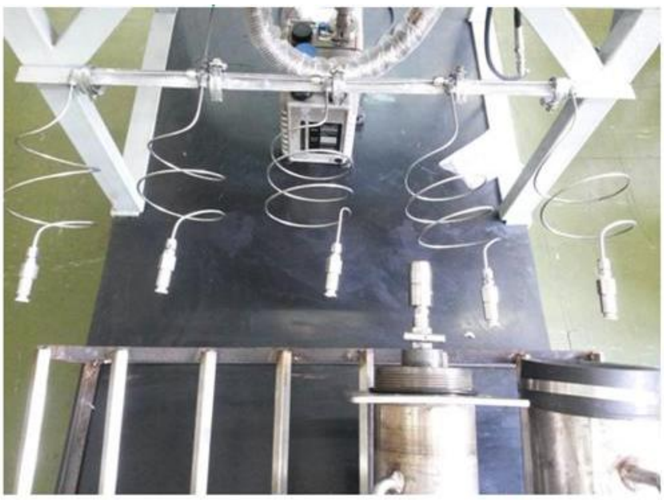

Figure 7: AISI 304 stainless steel tubing with 3.175 mm (1/8") inner diameter (inferior level) [9].

e) Needle valves, ringed connection, fast lock connectors, manovacuometer and AISI 304 stainless steel tubing for the radioactive gas transfer line setup (Fig.8) [9];

Figure 8: Needle valves, ringed connection, fast lock connectors, manovacuometer and AISI 304 stainless steel tubing for the radioactive gas transfer line setup [9].

f) Liquid nitrogen based freezing system (dewar) for gas liquefaction (Fig.9) [9];

Figure 9: Liquid nitrogen based freezing system for gas liquefaction [9].

g) Dewar with 11 cm thick walls lead shielding for the liquefaction (Fig.10) [9];

Figure 10: Lead shielding for the liquefaction dewar [9].

h) Acrylic box to isolate the transfer lines with air exit to be connected with the IEA-R1 Nuclear Reactor exhaust system (Fig.11) [9];

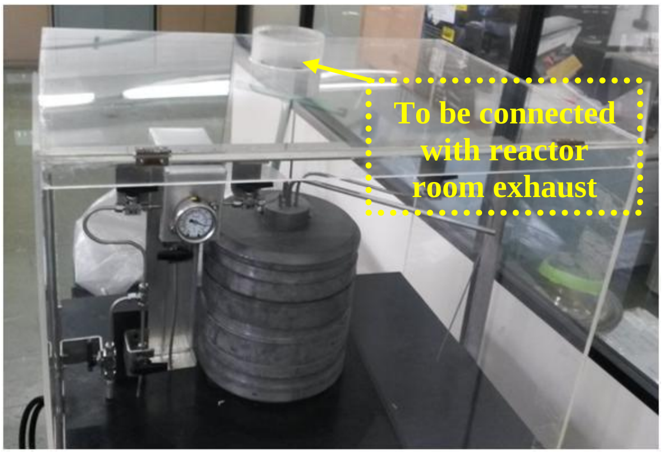

Figure11: Acrylic box with inner air exit to be connected with IEA-R1 Nuclear Reactor exhaust system [9].

i) Vacuum system with the mechanical vacuum pump [9]; and

j) Cylinder for storage and transportation 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 cm³ of inner volume (Fig.12) [9].

Figure 12: Cylinder for storage and transportation of the gaseous radiotracers [9].

In the first experimental production with the (IS), it was obtained an estimated activity of the $1.07x10^{11}$Bq (2.9 Ci) of ^{41}Ar distributed in two (STCs) utilizing the IEA-R1 Research Reactor with 4.5 MW and average thermal neutron flux of $4.71x10^{13}$ $n.cm^{-2}.s^{-1}$. However, the system has capacity for five storage and transport cylinders and the estimated maximum activity to be obtained is $7.4x10^{11}$Bq (20 Ci) per irradiation cycle. During this experimental irradiation, exposure rates were determined in: a) the lead shielding wall (10 mSv/h), in which the liquefied radioactive gas was concentrated; and b) the (STCs) after ^{41}Ar was transferred (25 mSv/h), by means of a portable radiation meter Teletector ® Probe 6150 AD-t/H [9].

1.2 Principle ALARA (As Low As Reasonably Achievable)

Justified use of radioactive material or radiation-emitting equipment implies observing the principles of occupational radioprotection, and in one of these, the principle of optimization, known as ALARA (As Low As Reasonably Achievable), establishes that in a project of installations that processing or use of radioactive materials or radiation emitting equipment, all exposures should be kept as low as reasonably practicable. Low dose epidemiological and radiobiological studies have shown that there is no dose threshold for stochastic effects, any exposure of a living tissue involves a carcinogenic risk depending of the radiosensitivity of that tissue per unit dose equivalent (somatic risk factor) or gonads, which may lead to genetic detriment in the descendants of the exposed individual [10,11].

2. MATERIALS AND METHODS

In the development of the automated irradiation system it will be used the infrastructure of the Radiation Technology Center and IEA-R1 Nuclear Research Reactor, both at IPEN-CNEN/SP. It will be based on several an innovative studies of industrial process control and production systems, and the use of Programmable Logic Controllers (PLC) and supervisory software, considering the safety standards from the IAEA and CNEN to guide the project, including mechanical solutions, software and hardware for the automation and process parameters studies.

2.1 System's production process review

Currently Irradiation System has its components structured according to the Fig.13. Operational routines are divided in four steps, as in shown Fig.14. This configuration and the operational routine will be analyzed to find out the best development of automated equipment for production of gaseous radiotracer.

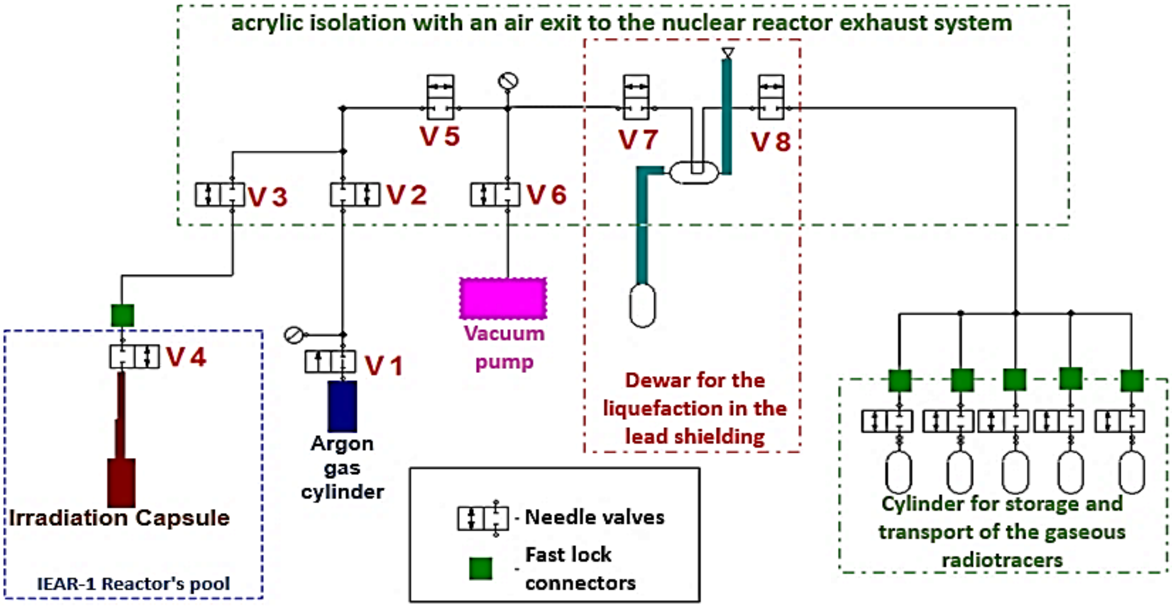

Figure 13: Operational structure of the irradiation system for gaseous radioisotopes production.

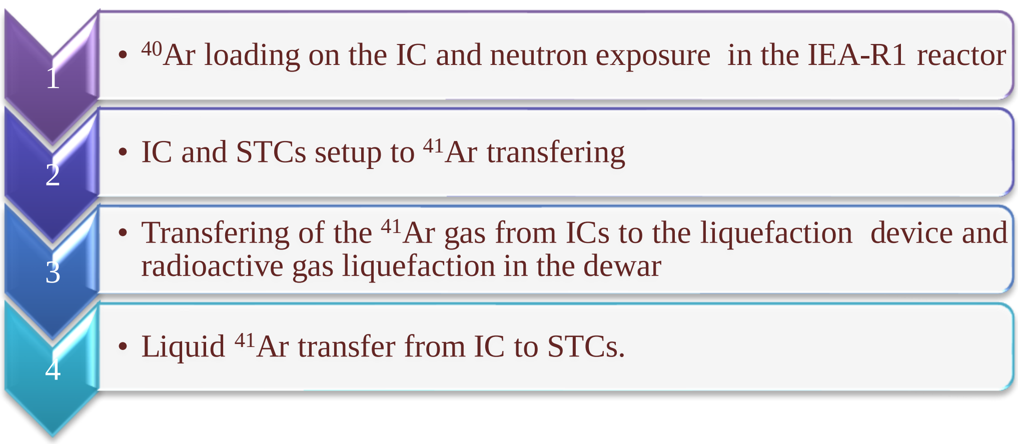

Figure 14: The argon-41 production proceeding steps [9].

2.2 Implementation of programmable logic controller and supervisory software

Programmable logic controller will be utilized in this study, because PLC is an equipment that can operate in aggressive industrial environments, and it provides user-mounted configurations as needed and advantages such as safety, reliability and easy maintenance. The National Electrical Manufacturers Association (NEMA) describes PLC as a digital electronic device that uses programmable memory for internal storage of instructions for specific actions, such as logic, sequencing, timing, counting and arithmetic, to control various types of machines and processes through input and output modules. It is made of interconnected electronic circuits, processors, memories, input and output devices and power supply. Its programming terminal can be compared to a computer specially adapted for industrial applications. For PLC implementation, supervision software should be configured based on the review of irradiation system components and operational process, because there is nothing commercialized to be directly implemented for this innovative IS [12].

3. CONCLUSIONS

In summary, the initial proposal evidenced in this research work, theme of a future PhD Thesis to develop an automated irradiation system for production of gaseous radioisotopes, controlled by a Programmable Logic Controllers (PLC), software development and the integration of current components brings an innovative configuration for this equipment allowing a remote control and consequently better security conditions, supplying gaseous radioisotopes (^{41}Ar and ^{79}Kr) in continuous scale, which can be used in industrial applications of emission tomography and flow measurement in petrochemical companies.

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