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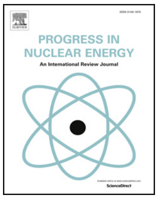

Irradiation system for production of gaseous radioisotopes used as tracers in industrial process measurements

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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} $\text{ncm}^{-2}\text{s}^{-1}$ 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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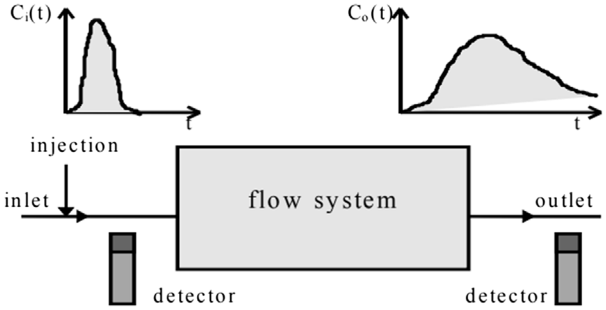

Fig. 1. RTD principle (IAEA, 2001).

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).

$$ {}_Z X^A + {}_0 n^1 \rightarrow [{}_Z X^{A+1}] \rightarrow ({}_Z X^{A+1})^* \rightarrow \gamma_{prompt} \tag{1} $$

$$ {}^{40}Ar + n \rightarrow {}^{41}Ar + \gamma_{prompt} \tag{2} $$

$$ {}^{78}Kr + n \rightarrow {}^{79}Kr + \gamma_{prompt} \tag{3} $$

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.

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

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).

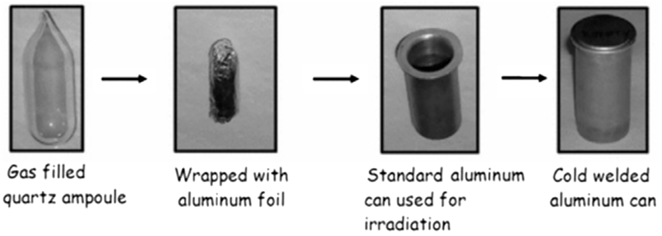

Fig. 2. Gaseous radioisotope production in quartz ampoules (Yelgaonkar et al., 2007).

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