Applied Radiation and Isotopes 116 (2016) 110–119

Contents lists available at ScienceDirect

Applied Radiation and Isotopes

journal homepage: www.elsevier.com/locate/apradiso

Production of gaseous radiotracers for industrial applications

V.K. Sharma [a], H.J. Pant [a,*], Sunil Goswami [a], K.C. Jagadeesan [a], S. Anand [b], S. Chitra [b], Y.S. Rana [b], Archana Sharma [c], Tej Singh [c], H.G. Gujar [c], Ashutosh Dash [a]

[a] Isotope Production and Applications Division, Bhabha Atomic Research Centre, Mumbai, 400085 India
[b] Health Physcis Division, Bhabha Atomic Research Centre, Mumbai, 400085 India
[c] Research Reactor Services Division, Bhabha Atomic Research Centre, Mumbai, 400085 India

HIGHLIGHTS

- Various prerequisite tests for irradiation of gaseous targets were performed.
- Procedure for production of gaseous radioisotopes was standardized.
- Detailed radiological safety analysis was carried out.
- Necessary approvals for production of gaseous radioisotopes were obtained.
- Production of gaseous radioisotopes began in DHRUVA reactor at Trombay, Mumbai, India.

ARTICLE INFO

Article history:
Received 1 July 2016
Accepted 12 July 2016
Available online 14 July 2016

Keywords:
Argon-41
Krypton-79
Radiotracer
DHRUVA
Tray rod facility
Quartz ampoule

ABSTRACT

This paper describes prerequisite tests, analysis and the procedure for irradiation of gaseous targets and production of gaseous radioisotopes i.e. argon-41 (^{41}Ar) and krypton-79 (^{79}Kr) in a 100 MW_{Th} DHRUVA reactor located at Bhabha Atomic Research Center (BARC), Trombay, Mumbai, India. The produced radioisotopes will be used as radiotracers for tracing gas phase in industrial process systems. Various details and prequalification tests required for irradiation of gaseous targets are discussed. The procedure for regular production of ^{41}Ar and ^{79}Kr, and assay of their activity were standardized. Theoretically estimated and experimentally produced amounts of activities of the two radioisotopes, irradiated at identical conditions, were compared and found to be in good agreement. Based on the various tests, radiological safety analysis and standardization of the irradiation procedure, necessary approval was obtained from the competent reactor operating and safety authorities for regular production of gaseous radiotracers in DHRUVA reactor.

1. Introduction

Artificially produced radioisotopes are widely used as radiotracers for troubleshooting, process diagnosis, measurements of process parameters and flow visualization in industrial process systems. The commonly carried out applications include leak detection in underground pipelines and heat exchangers, residence time distribution measurements in process systems, flow rate measurements in pipelines and canals, sediment transport investigations in ports and effluent/pollutant dispersion in aqueous systems (Gilath, 1977; Charlton, 1986; IAEA, 1990, 2004; Thyn et al., 2000; Pant and Yelgoankar, 2002; Pant et al., 2000, 2001, 2009a, 2009b, 2012, 2015; Pant et al., 2016). The selection of a radiotracer for a particular application depends upon physico-chemical compatibility, type of radiations emitted and their energies, half-life, radiochemical toxicity and activation cross-section for thermal neutrons. For tracing gaseous phase in industrial systems, gaseous radionuclide such as ^{41}Ar, ^{79}Kr, krypton-85 (^{85}Kr) and xenon-133 (^{133}Xe) are commonly used. Out of these gaseous radionuclides, ^{41}Ar and ^{79}Kr are the most suitable for use as radiotracers due to their favorable nuclear characteristics (Fries, 1977; Charlton, 1986; Thyn et al., 1988; Margrita and Sanlos-Gottin, 1988; Bernard et al., 1989; IAEA, 1990; Mercer et al., 2000).

Radioisotopes are produced by irradiating suitable target materials by thermal neutrons in a nuclear reactor for an appropriate time (IAEA, 2003). Physical form and characteristics of the target materials are very important for consideration of irradiation in a nuclear reactor. Targets which are non-explosive, non-pyrophoric, non-volatile, stable at irradiation conditions and do not contain impurities are used as targets for irradiation and production of

* Corresponding author.
E-mail addresses: hjpant@barc.gov.in, hjpant02@gmail.com (H.J. Pant).

http://dx.doi.org/10.1016/j.apradiso.2016.07.011
0969-8043/

radioisotopes. The production of any new radioisotope in a nuclear reactor requires detailed analysis and standardization of procedure before undertaking irradiation for the first time. The irradiation requirements and procedure are more stringent in case of gaseous targets due to associated handling problems such as its sealing in irradiation containers and high risk of radiological hazards that might arise due to leakage. The detailed experimental and theoretical analysis include qualification of irradiation containers, radiochemical and isotopic purity of the targets, rise in temperature, pressure, heat and reactivity during irradiation, possible accidental scenarios and their radiological impact, estimation of activity to be produced, optimization of irradiation parameters and obtaining necessary approvals from the competent operation and safety authorities for regularization of production.

At present, barring the production of gaseous radioisotopes in India, numerous other radioisotopes for various applications in healthcare, agriculture and industry are produced in a 100 MW (Th), heavy water moderated and cooled thermal research reactor DHRUVA having a maximum neutron flux level of 1.86×10^{14} neutrons $cm^{-2}\,s^{-1}$ (Sharma et al., 2002). In recent years, there arose a need to produce gaseous radioisotopes such as ^{41}Ar and ^{79}Kr for use as radiotracers in Indian industry. Therefore, this paper describes theoretical and experimental analysis of various aspects of irradiation for production of gaseous radioisotopes i.e. ^{41}Ar and ^{79}Kr in a high flux research reactor DHRUVA located at BARC, Trombay, Mumbai, India.

2. Production of gaseous radioisotopes

2.1. Irradiation containers

The irradiation container used for packing, sealing, encapsulating and irradiating target materials in a nuclear reactor should be made of high purity material that does not produce undesired radioisotopes. If produced, they should be short-lived. In addition to this, it should have good mechanical properties and should be stable at reactor operating conditions. In view of this, high-purity aluminum (Al) containers are used for packing and sealing target materials for irradiation in reactors. Usually solid targets are packed and sealed in standard Al-containers for irradiation, but gaseous targets need to be filled and sealed in quartz ampoules and subsequently encapsulated in the standard Al-containers as shown in Fig. 1. The container is capped with an aluminum lid and cold-welded. The Al-container was tested in the laboratory and was observed that it can withstand a pressure 10.2 kg/cm^2. The quartz is basically 99.995 W% silica (SiO_2) with alumina (Al_2O_3) as the major impurity. In addition to this, it also contains trace amount of sodium oxide (Na_2O), iron oxide (Fe_2O_3), calcium oxide (CaO), titanium oxide (TiO_2), potassium oxide (K_2O) and lithium oxide (Li_2O). These trace impurities do not produce their respective radioisotopes in significant quantities on irradiation in the reactor. Unique mechanical, chemical and thermal properties (high softening temperature and thermal resistance, low thermal expansion with high resistance to thermal shocks and high irradiation resistance) make quartz glass most suitable material for filling, sealing and irradiation of gaseous targets. In addition to this, it is easier to handle and break the quartz ampoules, and release the activity for further use as radiotracer. Some of the important properties of quartz glass are given in Table 1 (Bansal and Doremus, 2013). The photograph of the quartz ampoule and associated sequential steps used in filling and sealing the gaseous targets are shown in Fig. 1. The dimension of the quartz ampoule should be such that it is easily inserted and removed from the Al-container after it is wrapped in an aluminum foil. The wrapping of ampoules in aluminum foil before inserting into the Al-containers provides adequate cushioning against any breakage of the ampoules during handling.

Table 1
Physical and mechanical properties of quartz glass (Bansal and Doremus, 2013).

Property	Value
Density	2.2 g/cm^3
Hardness	5.5–6.5 Moh's scale
Design tensile strength	492 kg/cm^2
Design compressive strength	$> 1.1 \times 10^4$ kg/cm^2
Softening point	1683 °C
Strain point	1120 °C
Annealing point	1215 °C
Coefficient of thermal expansion (20–320°C)	5.5×10^{-7} cm/cm °C
Thermal conductivity	3.3×10^{-3} g cal cm/cm^2 sec °C
Maximum working temperature	1000–1300 °C

2.2. Inactive gaseous targets (argon-40 and krypton-78)

Inactive gaseous targets of argon-40 and krypton-78 were chosen for irradiation and production of ^{41}Ar and ^{79}Kr, respectively as these radioisotopes have suitable nuclear characteristics and can be used for tracing gas phases in industry. The nuclear characteristics of ^{41}Ar and ^{79}Kr are given in Table 2. Natural argon has

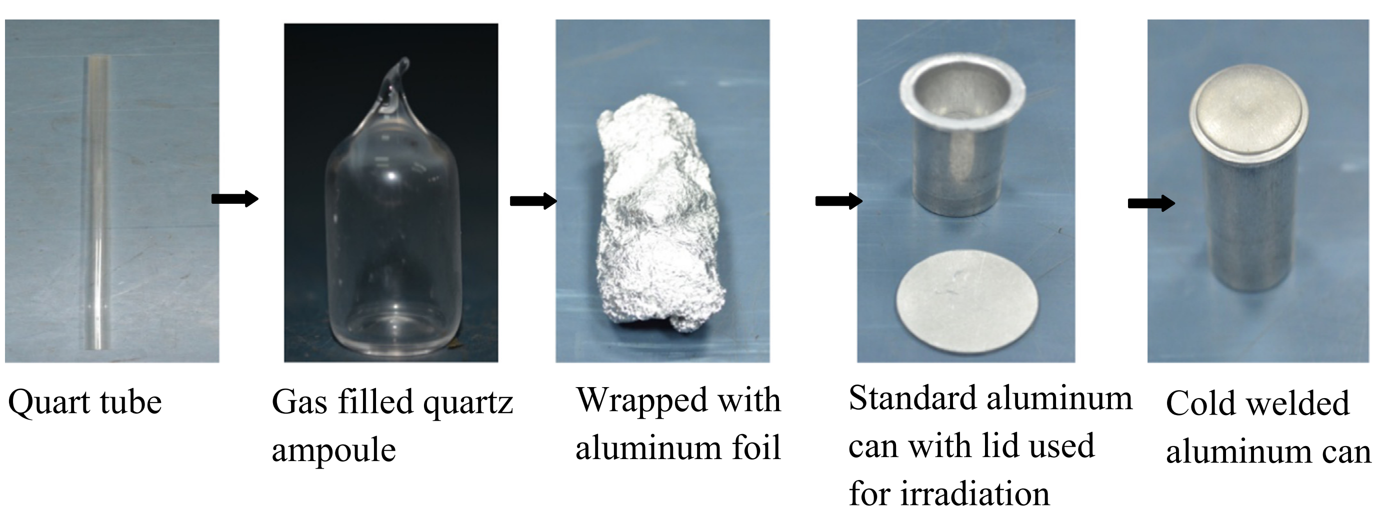

Fig. 1. Steps involved in sealing quartz ampoule in an irradiation container.