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Original Paper

Experimental measurement and Monte Carlo assessment of Argon-41 production in a PET cyclotron facility

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ABSTRACT

In a medical cyclotron facility, ^{41}Ar ($t_{1/2}$ = 109.34 m) is produced by the activation of air due to the neutron flux during irradiation, according to the $^{40}Ar(n,\gamma)^{41}Ar$ reaction; this is particularly relevant in widely diffused high beam current cyclotrons for the production of PET radionuclides. While theoretical estimations of the ^{41}Ar production have been published, no data are available on direct experimental measurements for a biomedical cyclotron. In this work, we describe a sampling methodology and report the results of an extensive measurement campaign. Furthermore, the experimental results are compared with Monte Carlo simulations performed with the FLUKA code. To measure ^{41}Ar activity, air samples were taken inside the cyclotron bunker in sealed Marinelli beakers, during the routine production of ^{18}F with a 16.5 MeV GE-PETtrace cyclotron; this sampling thus reproduces a situation of absence of air changes. Samples analysis was performed in a gamma-ray spectrometry system equipped with HPGe detector. Monte Carlo assessment of the ^{41}Ar saturation yield was performed directly using the standard FLUKA score RESNUCLE, and off-line by the convolution of neutron fluence with cross section data. The average ^{41}Ar saturation yield per one liter of air of ^{41}Ar, measured in gamma-ray spectrometry, resulted to be 3.0 ± 0.6 Bq/μA*dm^3 while simulations gave a result of 6.9 ± 0.3 Bq/μA*dm^3 in the direct assessment and 6.92 ± 0.22 Bq/μA*dm^3 by the convolution neutron fluence-to-cross section.

Introduction

In nuclear medicine, research on advanced and innovative technological solutions is necessary to improve diagnostic and therapeutic procedures, while concurrently assuring safety and protection of patients, workers, and the general population. This assumes particular relevance in the use of particle accelerators in the medical field, since in this case also the environment and the population living in areas adjacent to the site might be involved in an accident, as long as planned and unplanned releases of radioactivity.

Cyclotrons are used in nuclear medicine to produce short-lived radionuclides of biomedical interest, especially for Positron Emission Tomography, as well as in radiation oncology, hadron therapy. Knowledge of the radiation fields around these devices is necessary for the design of shielding, the classification of areas and the appropriate selection of safety systems; of specific relevance, is the assessment of the activation of the accelerator itself, building materials, and air during the routine use and the entire working life of the accelerator.

In particular, the production, and consequent release of radioactive gases in the external atmosphere is an important aspect of the radiation protection of workers and reference person (as defined by ICRP publication 103 [1], par. 193). In a medical cyclotron facility, ^{41}Ar ($t_{1/2}$ = 109.34 m) is the most important air activation product, due to the relatively high secondary neutron flux during irradiation and according to the $^{40}Ar(n,\gamma)^{41}Ar$ reaction (Fig. 1) [2].

In the literature, some analytical assessments of the ^{41}Ar production in air have been published, based on assumptions or experimental measurement of the neutron flux. A theoretical model of the production of ^{41}Ar in a cyclotron vault and its release in atmosphere were studied in several papers by Birattari et al. [3,4], where hypothesis on the shape and the energy distribution of the neutron field were done. Gutermuth et al. [5] and Biju et al. [6] performed FLUKA Monte Carlo assessment of ^{41}Ar concentration around proton accelerators in the 0.1–1 GeV energy range; in these works, Monte Carlo simulations were compared with results of analytical estimations, according to methods described in IAEA Report 283 [7] or NCRP Report 144 [8] respectively. An estimation of the production of ^{41}Ar due to photoneutrons near a 15 MV linear accelerator was performed by Chao et al. [9], based on experimental

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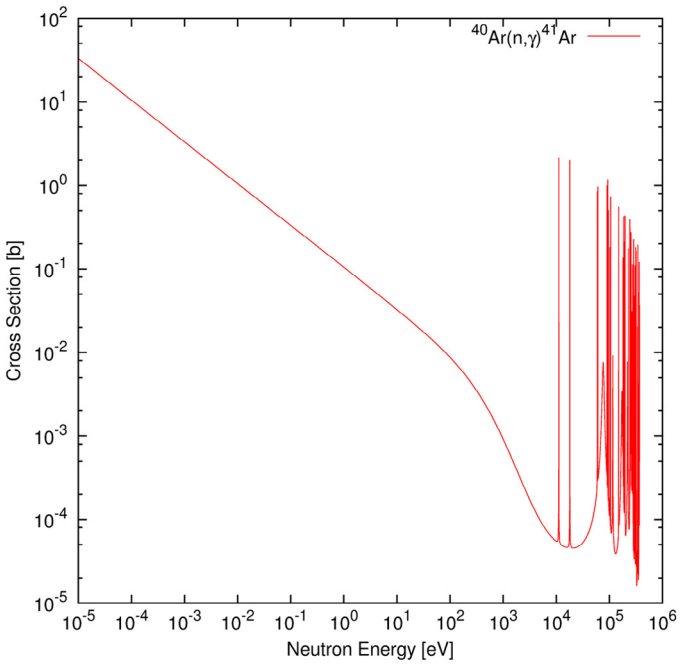

Figure 1. Cross section of the ^{40}Ar(n,γ)^{41}Ar reaction. This reaction has a non-negligible cross section for fast neutrons and a significant value of 660 mbarn at 0.025 eV [2].

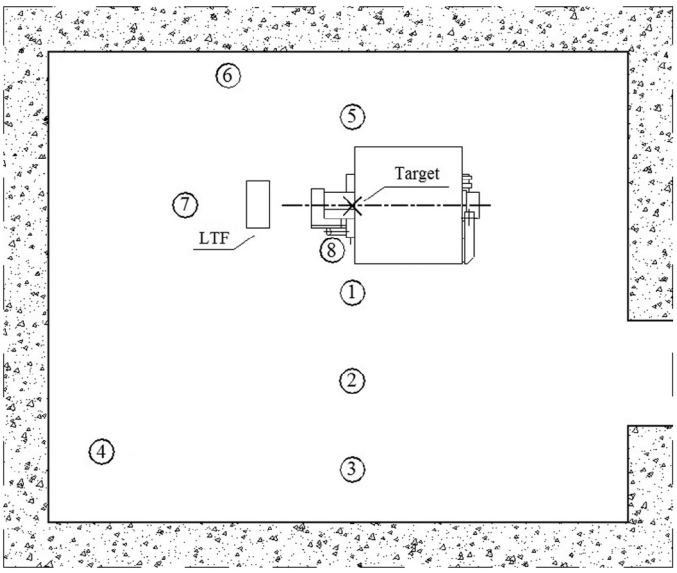

Figure 2. Sampling positions adopted during the measurement campaign within the bunker.

measurement of the neutron flux using Indium foils. Some experimental results are available as regards ^{41}Ar release from nuclear reactors [10,11]. Bezshyyko et al. [12] made an evaluation on the production of ^{41}Ar, based on analytical calculations and on a simplified Monte Carlo model of a self-shielded cyclotron, and finally Braccini et al. reported on experimental measurements of the induced radioactivity due to a 18 MeV proton beam directly extracted in air from a medical cyclotron [13]. However, no reports on direct measurement of ^{41}Ar production in air around biomedical cyclotrons are currently available.

In this work, we developed a sampling methodology for the measurement of ^{41}Ar inside the cyclotron vault. To compare the experimental measurements, a detailed Monte Carlo model of the bunker, the cyclotron and the target assembly, used in the University Hospital "S. Orsola-Malpighi", Bologna (Italy) has been created using the FLUKA code.

Materials and methods

Experimental setup

The cyclotron used in the irradiation tests and to which our simulations setup refers, is a PETtrace (GE Medical System), a compact cyclotron with vertical acceleration plane, capable of accelerating negative hydrogen and deuterium ions up to an energy of 16.5 and 8.4 MeV, respectively. The cyclotron is used for the routine production of Positron Emission Tomography (PET) radionuclides. The target system modeled was a GE assembly comprised of a silver chamber filled with [^{18}O]-water to produce Fluorine-18 by the (p,n) reaction. The front of the target body is sealed with a 25 μm Havar™ foil, an alloy of cobalt (42.5%), chromium (20%), nickel (20%) and traces of manganese, molybdenum, iron and others. The inner dimensions of the bunker are: 650 cm by 535 cm with a height of 350 cm and 200 cm thick concrete walls. Data on dimensions and characteristics of the bunker, the cyclotron and its components were taken from technical sheets and project drawings of the site. The ventilation rate inside the bunker is routinely fixed at 10 air changes per hour.

To measure the activity concentration of ^{41}Ar inside the cyclotron bunker, an extensive measurement campaign was performed: Marinelli beakers (typical total filling volume 1.36 dm^3) were placed inside the bunker, during routine productions of ^{18}F, in a series of marked positions, as reported in Fig. 2. Beakers 1, 2, 3, 4 were placed at a distance of 1, 2, 3, 4 m from the target respectively; beaker 5 was placed at 1 m while beakers 6 and 7 at 2 m from the target; beaker 8 was placed "in contact" with the target. Positions were marked on the floor, to be reproducible; all the beakers were placed at the same height (120 cm from the ground) as the target assembly used in the irradiation.

Marinelli beakers were sealed to simulate the absence of the ventilation, as in the Monte Carlo model described in the following; given its really small thickness, the influence of the plastic walls of the beaker can be neglected, and the activation of the air inside a beaker can be assumed to be equal to activation of air outside the beaker.

At the end of the irradiation, and after a waiting time of 10–20 minutes necessary to enter the bunker in safety conditions, the samples were removed, transferred to the gamma ray spectrometry laboratory (located on the same level at a distance of 25) and measured using an HPGe N-Type detector. The HPGe detector has a 30% relative efficiency and a resolution of 1.8 keV at 1332 keV. The high-resolution gamma-ray spectrometry system is based on digital electronics (Areva Canberra, distributed in Italy by TNE, Milan). The spectrometry system was calibrated in the 59–1836 keV range by means of a multi-radionuclide certified reference solution, obtained from an accredited Standardization Laboratory (Areva CERCA LEA, Pierrelatte Cedex, France). The calibration process was performed accordingly to the IEC 61452 standard [14], using the Genie 2000 software (Areva Canberra, distributed in Italy by TNE, Milan). A dual logarithmic polynomial efficiency curve was used. The method implemented in the software accounts for the propagation of the uncertainties in the calibration of the reference source (1–2% at 1 sigma level, depending on the peak in the mixture), in the tabulated yield (typically <1%), in the net peak area (<1% for calibration peaks) and in the interpolation of the curve (typically <3%). The calibration uncertainties therefore result of about 4–5% at 1 sigma level [15]. Samples were measured for 600–1800 seconds and all the