Frascati Physics Series Vol. 67 (2018)
6th Young Researcher Workshop 'Physics Challenges in the LHC Era'
May 7 and 10, 2018

VERIFYING THE PERFORMANCE OF THE ARGON-41 MONITORING SYSTEM FROM FLUORINE-18 PRODUCTION FOR MEDICAL APPLICATIONS

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Abstract

The aim of this work is to determine the detection efficiency of a Geiger–Muller detector placed in the terminal part of the chimney of the TR19PET cyclotron for environmental monitoring of ^{41}Ar emission (activity) through the chimney of the TR19PET cyclotron (19 MeV), installed at A. Gemelli University Hospital (Roma, IT) and routinely used in the production of positron-emitting radionuclides.

1 Introduction

In the medical field, cyclotrons are used both in diagnostics and therapy: in diagnostics they are used in the production of radioactive isotopes used, in particular, as tracers for Positron Emission Tomography (PET) [1]. The aim of this work is to characterize, from a radioprotection point of view, the emission of the radioactive gases following the production of the ^{18}F-FDG radiopharmaceutical by proton irradiation through the TR19PET cyclotron and to determine the detection efficiency of the Geiger–Muller detector through the use of Monte Carlo (MC) code MCNP. 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,γ)^{41}Ar reaction. This is particularly relevant in widely diffused high-beam current cyclotrons for the production of PET radionuclides, ^{18}F radionuclide in this case.

2 Cyclotron TR19PET

The TR19PET system is composed of cyclotron, cabinet of power supply and control system, cooling units and local shielding (in order to reduce at the source level the radiant beams emitted). The TR19PET cyclotron, produced by ACSI [2], is an isochronus compact and accelerates negative ions H^- in a plan vertically disposed up to a maximum energy of 19 MeV. ^{18}F-fluorine ion, the most used for PET in the form of ^{18}F-FDG radiopharmaceutical, is produced in water solution through the nuclear reaction ^{18}O(p,n)^{18}F. Once produced, the radionuclide (^{18}F) must be bound to a suitable molecule acting as a transporter, the FDG (Fluorodeoxyglucose) to create ^{18}F-FDG radiopharmaceutical, which accumulates in points of interest [3, 4, 1].

3 Problems following Fluorine-18 production

The main problems are due to: 1. The radionuclidic impurities of the irradiated (^{18}O) water, originated by the (^{18}F) FDG synthesis process [5]; 2. the ^{41}Ar ($t_{1/2}$ = 109.34 m) produced by the activation of air due to the neutron flux during irradiation, according to the ^{40}Ar(n,γ)^{41}Ar reaction [6]. The second problem is that the only exothermic reaction without threshold is the one wherein the neutron is captured by ^{40}Ar with the consequent production of ^{41}Ar. The natural concentration of argon-40 in air is 0.93% per volume. The argon-41 (half-life 109.2 minutes) emits β-radiation with a maximum energy of 1198 MeV and gamma rays of 1294 keV. The region of resonances is in the range between 10 keV and 15 MeV. In cyclotrons without their own shielding, most of the argon-41 inside the bunker air is produced by neutrons thermalized by the bunker concrete walls. The description with a 3D geometry of the FLUKA MC geometric model of the TR19 cyclotron, the cyclotron vault and the ducts through the vault walls (SimpleGeo and Flair) has been made by Infantino [7, 8]. Neutrons are generated with energy around 1 MeV and then slowed down to thermal energy with a peak around 4×10^{-7} MeV. Considering only the cross section value related to neutrons of energy 0.025 eV, the production of ^{41}Ar is understimated.

4 Geiger–Muller detector

The detector is placed within the internal wall of the terminal part of the cyclotron chimney with measure range from 0.1 μSv/h to 1 Sv/h[1] and energy range from 30 keV to >1 MeV. The cone geometry, as seen from the detector, has been used in order to estimate through the MCNP code the detector efficiency related to the activity of argon-41 in the cone. The following plume Gaussian model of dispersion in the environment has been used to describe the concentration in air C [MBq/m^3] at receptor level [5]:

$$C = \frac{Q}{2\pi\sigma_y\sigma_x\overline{u}} \exp\left\{-\frac{1}{2}\left[\frac{y}{2\sigma_y^2} + \frac{(z-h)^2}{2\sigma_z^2}\right]\right\}, \tag{1}$$

where x is the linear distance of the receptor, Q the emission rate [MBq/s], h the effective level of release, $\overline{u}$ the wind mean speed at release level, y the transverse distance of receptor from plume axis, z the receptor height at ground level, σ_y and σ_x the distribution standard deviations within the cloud along the trasverse and vertical directions, respectively. The measurements are evaluated assuming that the release effective height is equal to that of the chimney release (25 m), the receptor at ground level is located at variable distances of 100, 200, 300 m, the plant collocation is considered within an urban area. Finally, the wind evaluations are carried out at release level for a critical group of population, that is assumed to be located underwind, considering a costant wind direction in different climate conditions.

[1]The sievert (Sv) is a unit of ionizing radiation in the International System of Units, corresponding to 1 Sv= 1 J/kg.

5 Experimental measurements

The noble inert gas ^{41}Ar is only of local interest (sommersion) because of its rapid decay and it affects both skin (β particles) and internal organs (γ rays). The ^{41}Ar considered herein has a β electron-capture (EC) decay, a half-life $t_{1/2} = 1.8$ h and a decay constant $\lambda = 1.054\ 10^{-4} s^{-1}$. Moreover, the following data are considered: $A = 700$ Bq/μA (activity produced in air by ^{41}Ar), $I = 50\ \mu A$ (current intensity of the beam), $\lambda = 1.054\ \times 10^{-4} s^{-1}$ (decay constant). Two cases have been analysed herein: with and without air changes.

- Case 1: Without air changes
  Activity produced in air in Bq:

$$A_{produced} = 700\ \frac{\text{Bq}}{\mu\text{A}}\ 50\ \mu A = 35000\ \text{Bq}; \tag{2}$$

Number of nuclei produced:

$$N = \frac{A}{\lambda} = \frac{35000}{1.054 \times 10^{-4}} = 3.32 \times 10^{8}\text{Bq s}; \tag{3}$$

Maximum activity in 1 hour:

$$A_{max} = 35000 \times 3600 = 1.26 \times 10^{8}\ \text{Bq}. \tag{4}$$

Calculating the number of nuclei produced according to the law of radioactive decay, which, in addition to the decay term of the previous batch, also includes a second production term of the current batch, one obtains:

$$N = N_0 e^{-\lambda \Delta t} + \frac{A}{\lambda}\left(1 - e^{-\lambda \Delta t}\right)\ [\text{atomi}]. \tag{5}$$

Setting the time in intervals of 10 seconds each and considering

$$A = \lambda \times N\ [\text{Bq}], \tag{6}$$

one obtains the graph in Fig. 1.

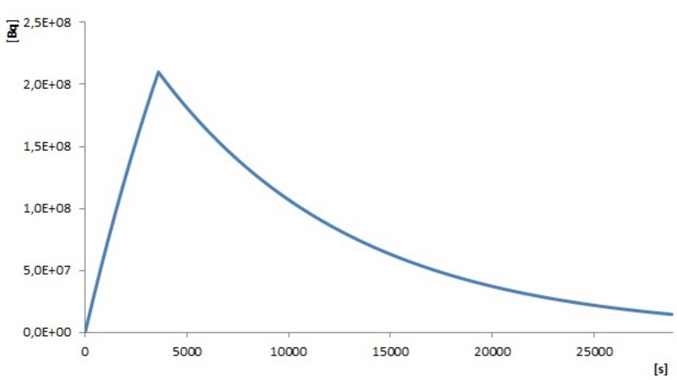


Figure 1: *Activity of* ^{41}Ar *vs. Time*

- Case 2: With air changes

  In this case the following data are considered: bunker volume $V_{cyclo} = 177.1$ m^3, branching ratio $b = 0.99157$, chimney diameter = 83 cm, area = 0.541 m^2, velocity during irradiation = 0.273 m/s, velocity during no irradiation = 0.100 m/s and irradiation time = 60 min. Moreover, a cone geometry has been considered, hypothesizing a height $h = 2$ m and a volume $V_{cone} = 1.08$ m^3, both seen by the detector:

$$Air\ changes\ rate\ (during\ irradiation) = \frac{2V_{cyclo}}{h} = 0.0148\ \frac{\mathrm{m}^3}{\mathrm{s}}, \tag{7}$$

$$Air\ changes\ rate\ (no\ irradiation) = \frac{a.c.r.\ (irr.)}{3} = 0.049\ \frac{\mathrm{m}^3}{\mathrm{s}}, \tag{8}$$

where *a.c.r.(irr.)* is the air-change rate during the irradiation.

The calculation of the detector efficiency through the use of MCNP, with cps standing for counts per second, proceeds along the following steps:

$$\epsilon = 4.72 \times 10^{-7}\ \frac{\mathrm{cps}}{\mathrm{Bq}}, \tag{9}$$

$$N_{expelled} = 2.77 \times 10^{7}\ \ [atomi]\,, \tag{10}$$

where the number of expelled and recycled nuclei is calculated as:

$$N_{expelled} = \left[N_0 \exp^{-\lambda\Delta t} + \frac{A}{\lambda}\left(1 - \exp^{-\lambda\Delta t}\right)\right] \times \frac{V_{air\ changes}}{V_{cyclo}} \times \Delta t, \tag{11}$$

$$N_{recycled} = N_0 - N_{expelled} = 3.32 \times 10^{9} - 2.77 \times 10^{7} = 3.3 \times 10^{9}\ [\mathrm{atomi}]\,. \tag{12}$$

Consequently, the expelled and recycled activities are calculated according to the following steps:

$$A_{expelled} = \lambda \times N_{expelled} = 3 \times 10^{3}\ [\mathrm{Bq}]\,, \tag{13}$$

$$A_{recycled} = \lambda \times N_{recycled} = 1.054 \times 10^{-4} \times 3.3 \times 10^{9} = 3.47 \times 10^{5}\ [\mathrm{Bq}]\,, \tag{14}$$

$$\frac{A_{expelled}}{air\ changes\ rate} = \frac{3 \times 10^{3}}{0.148} = 20270.3. \tag{15}$$

The activity potentially visible reads:

$$A_{potentially\ visible} = A_{expelled} \times \frac{v \times \Delta t}{H_{seen\ by\ detector}} = 27668.96\ \mathrm{Bq}. \tag{16}$$

Figure 2 presents the activity of ^{41}Ar inside the cyclotron (left) and the activity of the expelled ^{41}Ar (right) with respect to time:

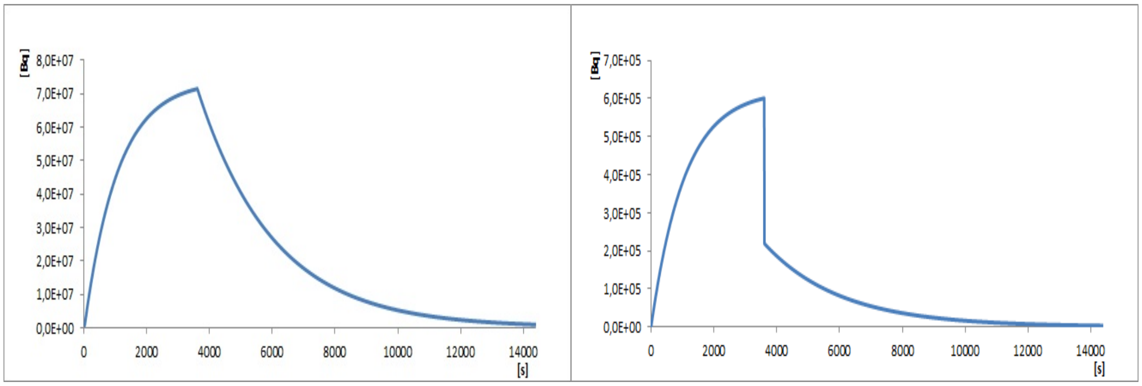
$$\frac{\mu Sv}{h} = \frac{1.3 \times 10^{-2}}{1939.129} \times 1000 = 6.68 \times 10^{-3} \frac{\mu Sv}{h}. \tag{17}$$

At last, the counting rate is given by:

$$R = \epsilon b A_{potentially\ visible} = 1.3 \times 10^{-2}. \tag{18}$$

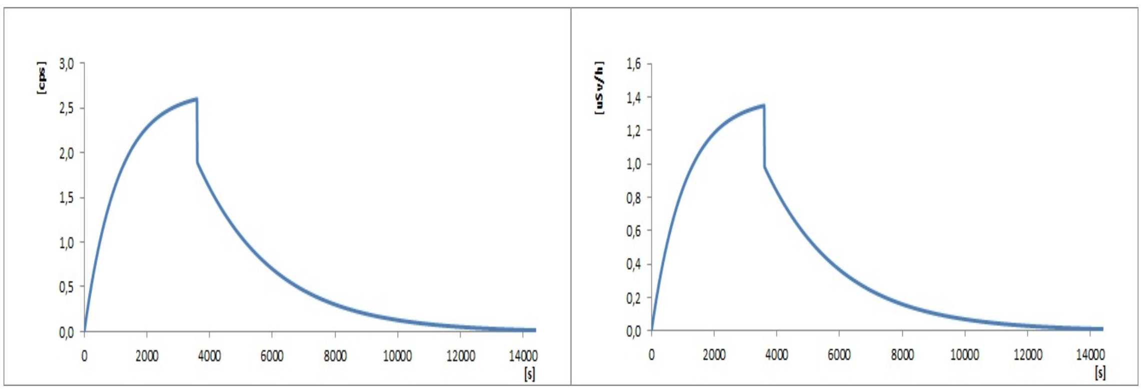
Figure 3 presents the counts per second (left) and the counting rate per hour ($\mu Sv/h$, right), as seen by the detector, as a function of time.

Finally, in Figure 4 one can see the optimized counts per second, after monitoring the emitted activity.



(a) Graph 1 (b) Graph 2

Figure 2: *Graph 1: Activity* ^{41}Ar *inside cyclotron vs. Time; Graph 2: Activity* ^{41}Ar *expelled vs. Time*



(a) Graph 1 (b) Graph 2

Figure 3: *Graph 1 cps seen by detector; Graph 2* $\mu Sv/h$ *seen by detector*

6 Conclusions

In conclusion, the detection efficiency determined through the use of Monte Carlo codes is very low (4.7 $\times 10^{-7}$ cps/Bq), and the counting rate of 1.3 $\times 10^{-2}$ is low as well. Consequently, in normal operating conditions, the detector does not produce any signal and gives a totally inefficient contribution related to its functions. Other monitoring systems are currently being studied by our research team in order to achieve a greater efficiency, finalized to the radiological impact assessment.

7 Acknowledgements

I would like to thank my collaborators Professor R. Remetti (”Sapienza University of Rome Italy, Department of Energetics) and Dr. L. Indovina (Physics Department, University Hospital ”A. Gemelli” Rome, Italy) for the work presented here and the organisers of the 6th Young Researchers Workshop: Physics Challenges in the LHC Era for the invitation.

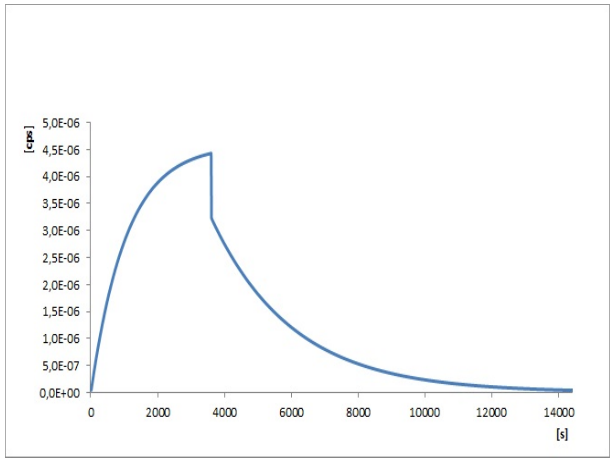


Figure 4: *Counting rate optimized monitoring activity emitted*

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