MONTE-CARLO SIMULATIONS ON RADIOLOGICAL CHARACTERISTICS OF A CYCLOTRON USED FOR PRODUCTION OF ^{18}F

G. Asova*, M. S. Yavahchova, N. Goutev, D. Tonev

Institute for nuclear research and nuclear energy, Bulgarian Academy of Sciences
1784, Sofia, Bulgaria
** corresponding author: gasova@inrne.bas.bg*

Abstract

Cyclotrons used for production of radioisotopes for nuclear medicine are already well established around the world. The Institute for Nuclear Research and Nuclear Energy is building such a cyclotron-based facility for production of and R&D on radioisotopes. Production of ^{18}F-based radiopharmaceuticals is the primary goal of the project.

^{18}F is obtained by bombarding a target filled with ^{18}O-enriched water with protons. The nuclear reaction used is ^{18}O (p, n) ^{18}F. During the irradiation strong fields of secondary neutrons and gamma rays are generated around the target. These fields induce nuclear reactions within the target, cyclotron components, beam lines, local shielding of targets, walls of the cyclotron vault that become radioactive as time goes on.

The aim of the present work is using numerical methods to evaluate the radioactive materials generated during the operation of the facility that are important for daily maintenance and decommissioning in future.

Key words: *cyclotron, 18F, Monte-Carlo simulations, radioactive waste*

Introduction

The Institute for Nuclear Research and Nuclear Energy at the Bulgarian Academy of Sciences is building a cyclotron facility for production of and R&D on radiopharmaceuticals. The facility is based on a TR-24 cyclotron capable to deliver proton beams with variable energy in the range between 15 and 24 MeV and beam currents up to 400 uA [1]. Such beam parameters give possibilities for production of various PET and SPECT isotopes. The major objective for INRNE is ^{18}F since the number of nuclear imaging procedures in oncology using ^{18}F in Bulgaria increases over the last few years [2, 3].

Building such an infrastructure requires detailed knowledge on the capabilities of the accelerator, the physics processes involved and the hazards related to them during or after the operation of the facility. Therefore, one of the tasks INRNE is working on at the moment is related to buildup and decay of radioactive waste during the production of radioisotopes. The buildup of radioactive material is due to activation by secondary particles, mainly neutrons, generated in nuclear reactions between the proton beam and the irradiated targets – ^{18}F is generated by a (p, n) reaction on ^{18}O-enriched H_2O-target. Attention is paid to the activation of components of the targets as they are the most activated. The radionuclides generated within the target vicinity including the cyclotron itself and the concrete walls of the vault housing the cyclotron are also evaluated.

Methodology of the numerical simulations, characterization of the secondary neutrons source

FLUKA [4, 5] has been used for the simulations due to its capabilities for numerical evaluation of emission and transport of secondary particles and, with some limitations, to evaluate the waste generated as a result of reactions related to secondary emission sources.

The first step of the Monte-Carlo simulations was to cross check the simulation parameters against experimental data. This has been done using a model of a real target for production of ^{18}F and various sets of beam parameters [6]. The target – Fig. 1a, consists of a volume for the $[^{18}O]H_2O$, shown with light green, where the $^{18}O(p, n)\,^{18}F$ reaction takes place, and a body, both covered by Havar foil. Afterwards, assuming most intense parameters of the impinging proton beam, i.e. highest beam energy and current, the target has been irradiated for six hours and the particles, emitted as a result of the (p, x) reactions with the body of the target, have been stored on disk to be used later as a source of secondary particles for irradiating all components within the cyclotron vault. Here *x* denotes neutrons and gamma rays as they are of practical interest

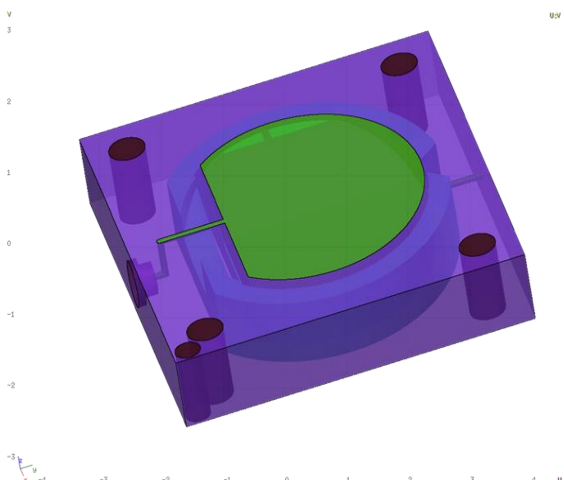
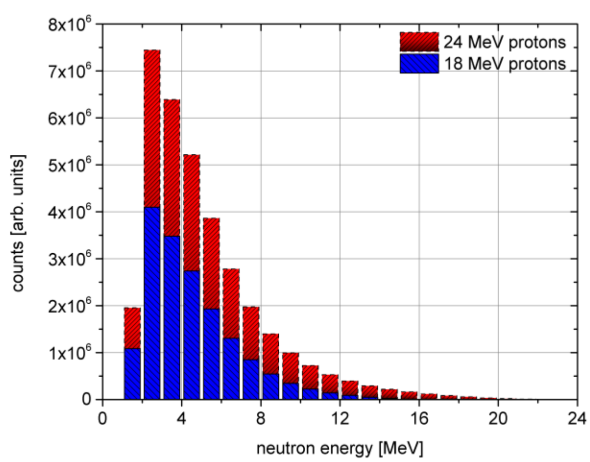


Figure 1a. Model of a high-current liquid target for ^{18}F production. In green is the liquid volume. The proton beam impinges orthogonally to the target front surface.

Figure 1b. Spectrum of neutrons emitted



from the liquid water volume at proton beam energies of 18 and 24 MeV

when dealing with proton accelerators at these energies [7]. The neutrons and gamma ray spectra have already been discussed [6]. As shown with red bars in Fig. 1, the energy of the neutrons covers the full range of energy up to the energy of the primary particles corrected with the Q-value of the $^{18}O(p, n)^{18}F$ reaction as the majority are low energy to fast ones. The same trend is seen also for 18 MeV protons.

Radionuclides generated in the foil

The target and more specifically the foil separating the liquid volume, where the nuclear reaction takes place, from the vacuum vessel of the transport beamline is one of the most often handled activated components. The 30 um thick Havar foil needs to be exchanged every 5000 uA/h. Table 1 shows some long-living radionuclides generated in the foil and the corresponding activities after one irradiation session of 6 hours at 24 MeV and 400 uA. Some of the radionuclides exceed the local exemption limits, denoted in bold, after only one session thus the foils need to be handled and stored as radioactive waste.

Table 1. Radionuclides generated in the Havar foil after one irradiation session of six hours at 24 MeV and 400 uA. In bold are exemption limits according to [8].

Nuclide	Half life	Activity / **exemption limit**
^{185}W	75 d	5.6 MBq
^{181}W	121 d	7 MBq
^{96}Tc	4.3 d	126 MBq / **1 MBq**
^{93}Mo	4e3 y	0.6 Bq
^{59}Fe	44 d	558 MBq
^{58}Co	71 d	2.7 MBq / **1 MBq**
^{56}Mn	2.6 h	18MBq / **0.1 MBq**
^{56}Co	77 d	0.1 MBq/ **0.1 MBq**
^{55}Fe	2.7 y	0.03 MBq / **1 MBq**
^{54}Mn	312 d	0.7 MBq
^{52}Mn	5 d	71 MBq / **0.1 MBq**
^{51}Cr	28 d	23 MBq / **10 MBq**
^{3}H	12.3 y	0.09 GBq / **1 GBq**

Vault geometry

The TR-24 cyclotron will be installed in a special vault for radiation protection. A scaled view of the vault geometry is shown in Fig. 2. For the simulations the target for ^{18}F production is positioned either in p. F or in p. M. The flux of secondary particles, neutrons and gamma rays, and the products of activation are evaluated for both locations.

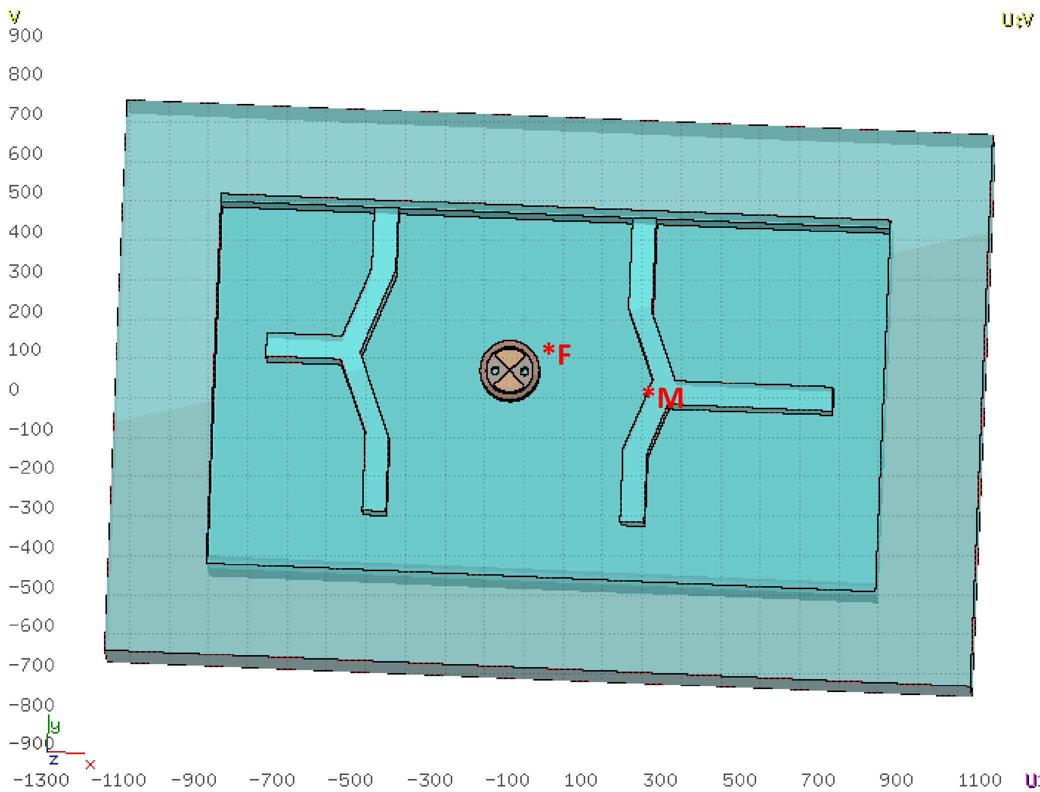


Figure 2. Top view of the vault with the cyclotron in the central part. The target for production of ^{18}F is located at positions F or M. The axes dimensions are in centimeters.

Radionuclides in the cyclotron and the concrete walls

Figure 3 shows the radionuclides generated in the cyclotron iron parts for the two target locations from Fig. 2 – on the left position F is chosen, position M is on the right. No additional neutron or gamma shielding is assumed around the target port. The operation time is limited to a month with five working days per week, six hours of irradiation time per day, 400 uA at 24 MeV. The activities shown are scored after a month of cooling time. It can be seen that choosing position M as a target location, away from the cyclotron body, leads to lower activities of ^{55}Fe, ^{59}Fe and ^{54}Mn, thus making the operation of the cyclotron safer for the personnel and simplifying its decommissioning.

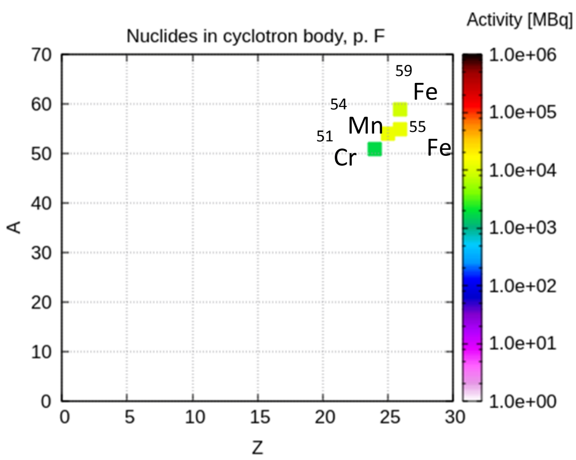


Figure 3a. Radionuclides in the cyclotron body for the target at position F.

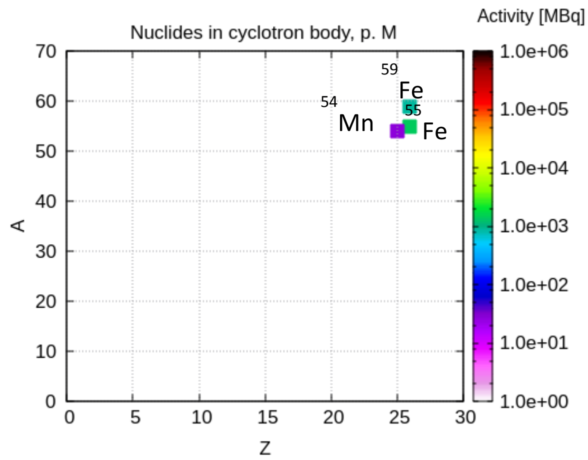


Figure 3b. Radionuclides in the cyclotron body for the target at position M.

Figure 4 shows the radionuclides generated in the innermost 20 cm of the walls of the vault for the two target positions from Fig. 2 – F and M on the left and right side correspondingly. Again

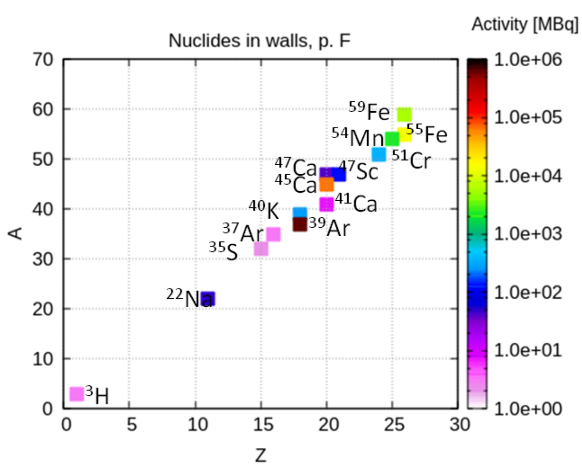


Figure 4a. Radionuclides for the target at position F.

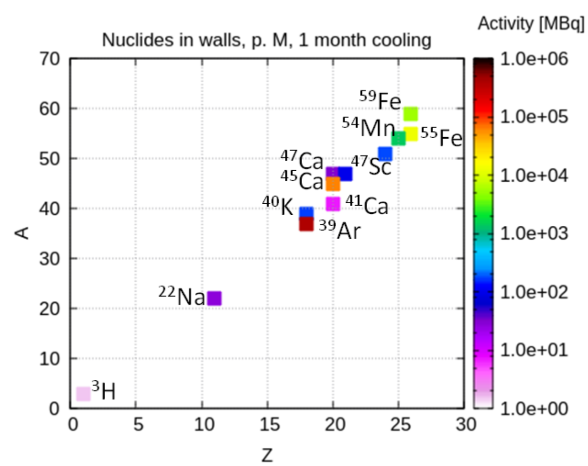


Figure 4b. Radionuclides for the target at position M.

Figure 4. Radionuclides in the innermost 20 cm of the walls for two target locations. The cyclotron has been operated for a month and additional time of 1 month is used for cooling.

the same irradiation conditions are used with additional cooling time of a month. The walls are made of standard Portland cement, containing H, Na, O, K, Al, Si, Ca, Fe, density 2.42 g cm^{-3}. ^{35}S ($T_{1/2}$ = 87.5 d) and ^{37}Ar ($T_{1/2}$ = 35 d) are generated only for target location F with possible parent nuclei ^{35}S from (n, γ) and ^{40}Ca from (n, $\alpha\gamma$) reactions correspondingly. For both target positions ^{39}Ar has activities above the exemption limits meaning this concrete layer has to be stored in specific conditions if the cyclotron facility would be running for a month with the irradiation scheme mentioned above and no additional shielding around the target.

Conclusions

This work shows the first numerical estimation of the radionuclides generated in the vicinity of a target for production of ^{18}F. The target is not shielded locally. Only reactions based on activation by secondary neutrons generated within the target have been considered. Two target locations have been studied showing that positions away from the cyclotron are preferable with respect to activation of the cyclotron itself and a surface layer of 20 cm of the concrete walls. An additional local target shielding will be considered in order to decrease the activities of long-living radionuclides in the walls like ^{35}Sc, ^{39}Ar, ^{54}Mn, ^{55}Fe, etc.

Acknowledgements

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