Status of Simulations for the Cyclotron Laboratory at the Institute for Nuclear Research and Nuclear Energy

G Asova, N Goutev, D Tonev, A Artinyan
Institute for Nuclear Research and Nuclear Energy, BAS, BG-1784 Sofia, Bulgaria
E-mail: gasova@inrne.bas.bg

Abstract. The Institute for Nuclear Research and Nuclear Energy is preparing to operate a high-power cyclotron for production of radioisotopes for nuclear medicine, research in radiochemistry, radiobiology, nuclear physics, solid state physics. The cyclotron is a TR24 produced by ASCI, Canada, capable to deliver proton beams in the energy range of 15 to 24 MeV with current as high as 400 µA. Multiple extraction lines can be fed. The primary goal of the project is the production of PET and SPECT isotopes as $^{18}$F, $^{67,68}$Ga, $^{99m}$Tc, etc.

This contribution reports the status of the project. Design considerations for the cyclotron vault will be discussed for some of the target radioisotopes.

1. Introduction
The Institute for Nuclear Research and Nuclear Energy (INRNE) at the Bulgarian Academy of Sciences is building a facility dedicated to: production of well-established medical radioisotopes; research and development on emerging radioisotopes for nuclear medicine; fundamental and applied research with radiotracers in areas related to life sciences and industry [1]. The facility is based on a TR24 cyclotron capable to deliver proton beams with current up to 400 µA and energy as high as 24 MeV. The cyclotron has already been produced by ASCI and delivered at the INRNE with its ancillary equipment like RF and water-cooling systems, a single Y-beamline equipped with steering and focusing magnets, etc. The construction of the building housing the cyclotron vault and the radiochemistry laboratories still has to be done. Meanwhile efforts are dedicated to numerical studies on the possibilities to produce various isotopes for medicine and radiological characterization of the setup.

The major objective for the start-up is the widely used $^{18}$F–FDG produced via $^{18}$O (p, n) $^{18}$F reaction in $^{18}$O-enriched target volume and, therefore, the radiological characterization has to take into account production of neutrons and γ-rays. Part of the neutrons result from the nuclear reaction within the target volume, whereas another part from primary particles (protons) or secondaries (neutrons, γ-rays, etc.) interacting with cyclotron components, target, walls of the facility. γ-rays can be generated by ongoing $^{18}$F–decay during irradiation or by neutron-induced reaction with various components. This study shows numerical estimations of some internal hazards coming along with the operation of the facility at maximal beam power. Radioactive materials generated in the $^{18}$F–target and the vault will be discussed.
2. Considerations for radiological characterization

2.1. Methodology for the simulations
For the simulations shown here FLUKA [2, 3] has been used due to its capabilities to numerically estimate emission and transport of secondary particles and to evaluate the radioactive waste generated as a result of reactions related to the secondary emission sources.

2.2. Radiation source
To adjust the simulation settings a real-size target for $^{18}$F was initially irradiated using various sets of beam parameters [4]. Despite the fact that the highest beam current and energy are not needed to be used for $^{18}$F production, afterward such were chosen to irradiate the target due to the stringent conditions they would impose on the radiation environment [5]. The target was irradiated for six hours and the particles, emitted in a spherical volume as a result of the $(p, x)$ reactions with any of the target components, have been collected to be used later as sources irradiating further components. Here x denotes neutrons and $\gamma$-rays as they are of practical interest when dealing with proton accelerators [5]. 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 ($Q = -2.44$ MeV) and the energy lost on a Havar foil, separating the area of the liquid target from the vacuum chamber of the setup. Majority of the neutrons are low energy to fast ones. The same trend is seen also for energy of 18 MeV.

The simulated spectrum for 18 MeV and 100 $\mu$A results in fluence rate of about 2.5e11 n/cm$^2$/s which can be compared to the measurement results shown in [6] where 2.4e10 n/cm$^2$/s are given for 15 $\mu$A and 17 MeV. The difference of an order of magnitude might be due to the lower beam current used in the measured data.

Figure 2 shows the fluence of secondary neutrons overlaid on the target geometry.

2.3. Fields of secondary particles within the cyclotron vault
The vault is designed to house the cyclotron and four irradiation areas as only one of them will be used for $^{18}$F production. There are two possible locations for the $^{18}$F–target in the vault - close to the cyclotron but still externally to it and in a separated irradiation room. Figure 3 shows the distribution of neutrons when using both positions - the highest density shown in black is the exact center of mass of the source from Section 2.2. The source obtained in the step described above is positioned in either location of the target and the particles are transported further depending on their spatial coordinates and momentum vector components in the source.

![Figure 1. Spectrum of neutrons emitted from the liquid water volume at proton beam energies of 18 and 24 MeV and current of 400 $\mu$A. Here the proton beam meets a Havar foil separating the enriched water from the vacuum chamber at normal angle of incidence.](image)
Figure 2. Fluence (particles per cm\(^2\) per primary particle) of neutrons crossing the target body in the three planes. Proton beam with energy of 24MeV and current of 400 \(\mu\)A impinges into the \((x, y)\) plane.

(a) Neutrons in the vaults when positioning the target within the vault inner area.

(b) Neutrons in the vaults when positioning the target outside the vault inner area in a dedicated irradiation room.

Figure 3. Top view of the neutron fields in the cyclotron vault for two different positions of the liquid target. The cyclotron is shown with a black circle in the center of the bunker. Thinner concrete walls separate the cyclotron area from the four irradiation areas.

phase space. The vault space is surrounded by 2.5 m thick walls made of high-density concrete as shown in Fig. 3. The target area is not shielded additionally. The neutron distribution homogeneity around the target is disturbed by cyclotron metal parts mostly in Fig. 3(a), inner and outer shield walls (Fig. 3(b)). As it might be assumed that the areas outside the vault will be used for chemical synthesis, target preparation, etc., it is desirable to minimize any leakages, thus, making the position of the target shown with Fig. 3(b) preferable if no further shielding is used. Additionally, the shielding can be improved as it is shown with Fig. 4 where a few centimeters thick marble-enriched concrete is used locally around the target. Further optimization of the thickness of this shielding and its chemical composition still has to be done.

3. Activation of the vault walls

The distribution shown in Fig. 2 can be used to study the activation of components of the target itself. It has already been shown [4, 7] that the nuclides generated in the Havar foil contain a
number of long-living ones ($^{3}$H, $^{58}$Co). This study focuses on the activation of the vault using the distributions from Fig. 2 and Fig. 3. It is assumed that a sacrificial layer takes 20 cm from the wall thickness including also the walls, separating the cyclotron from the irradiation areas. The last ones have thickness of 60 cm. Table 1 shows some of the nuclides generated in the sacrificial layer after a month of operation with daily irradiation conditions as above - 6 hours at 24 MeV and 400 $\mu$A, immediately after End of Beam (EOB) and after 3 weeks of cooling time.

Table 1. Some of the nuclides generated in the outmost 20 cm of the walls of the cyclotron vault. The irradiating target is positioned as in Fig. 3(b).

| Isotope   | Activity at EOB [Bq] | Activity in 4 weeks [Bq] | Parent nucleus |
|-----------|----------------------|--------------------------|----------------|
| $^{59}$Fe | $2.1 \times 10^{10}$  | $1.4 \times 10^{10}$     | $^{58}$Fe       |
| $^{56}$Mn | $1.8 \times 10^{9}$   | -                        | $^{56}$Fe       |
| $^{55}$Fe | $1.8 \times 10^{10}$  | $1.7 \times 10^{10}$     | $^{55}$Mn       |
| $^{54}$Mn | $4.3 \times 10^{9}$   | $4.1 \times 10^{9}$      | $^{54}$Cr, $^{54}$Fe |
| $^{51}$Cr | $1.1 \times 10^{9}$   | $5.7 \times 10^{8}$      | $^{52}$Cr       |
| $^{47}$Ca | $1.1 \times 10^{9}$   | $1.6 \times 10^{7}$      | $^{46}$Ca       |
| $^{47}$Sc | $1.0 \times 10^{9}$   | $5.0 \times 10^{7}$      | $^{47}$Ti       |
| $^{45}$Ca | $2.3 \times 10^{10}$  | $2.0 \times 10^{10}$     | $^{44}$Ca       |
| $^{41}$Ca | $2.5 \times 10^{6}$   | $2.5 \times 10^{6}$      | $^{40}$Ca       |
| $^{42}$K  | $2.2 \times 10^{11}$  | -                        | $^{41}$K        |
| $^{41}$Ar | $5.9 \times 10^{6}$   | -                        | $^{40}$Ar       |
| $^{39}$Ar | $8.1 \times 10^{7}$   | $8.1 \times 10^{7}$      | $^{39}$K        |
| $^{37}$Ar | $5.2 \times 10^{11}$  | $2.9 \times 10^{11}$     | $^{36}$Cl       |
| $^{36}$Cl | $2.8 \times 10^{4}$   | $2.8 \times 10^{4}$      | $^{39}$K        |
| $^{24}$Na | $1.7 \times 10^{12}$  | -                        | $^{27}$Al       |
| $^{14}$C  | $2.2 \times 10^{3}$   | $2.2 \times 10^{3}$      | $^{13}$C        |
| $^{3}$H   | $3.1 \times 10^{6}$   | $3.1 \times 10^{6}$      | (p, $^{3}$H)    |

$^{41}$Ar resulting from neutron capture on $^{40}$Ar is also seen in the air of the vault in activities in the range of $1 \times 10^{7}$ Bq - above the limit of 200 Bq/m$^3$ with respect to the volume of the vault increasing the cooling time before maintenance access. In 18 hours the activity decreases to $3 \times 10^{4}$ Bq with which the specific activity within the inner cyclotron room decreases below.
the exemption limit. Further estimations are needed considering that the usable lifetime of a cyclotron can be about 20 years. At the moment such estimations with the software package used can be done for operation time of one month.

4. Conclusion
The distribution of neutrons emitted as a result of primary reaction on $^{18}$F target from a mid-energy high-intensity cyclotron was evaluated using Monte-Carlo simulations. Two different locations of the target were considered as the one positioned in a separate irradiation room showed to be preferable with respect to possible leakages of low-energy neutrons through the walls of the vault. It was also shown that local target shielding with marble-enriched concrete would lead to further improvements.

A number of toxic and long-living isotopes were seen in the sacrificial layer of the inner walls ($^{55}$Fe, $^{45}$Ca, $^{41}$Ca, $^{39}$Ar). $^{41}$Ar is also seen in the air limiting the time needed for maintenance access.

5. Future steps
Characterization of the vault depends on the targets being irradiated and therefore more isotopes will be studied. At the moment efforts are spent on the production routes of $^{67,68}$Ge and $^{99m}$Tc and they will also be used to assess the activation in the vault. Additionally the local target shielding needs to be optimized with respect to ease of usage (weight), composition, leakage and neutrons and $\gamma$-rays.

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