

Local Shielding of Target for Production of ^{18}F – Numerical Optimization

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Abstract. The results presented in the current paper are part of the studies which we are conducting in regards with the radiological characterization of the cyclotron facility that is currently being built by the Institute for Nuclear Research and Nuclear Energy at the Bulgarian Academy of Sciences. The facility is going to be dedicated to research and development of radiopharmaceuticals, and ^{18}F is of primary interest.

Adding a local shielding around the target will be useful in limiting the spatial distribution of the emitted secondary neutrons (during the target irradiation) and the activation of the bunker walls. The effect of adding a layer of borated polyethylene is studied. We are considering a simplified spherical geometry, divided into spherical shells, of the bunker walls. By employing the FLUKA Monte-Carlo transport code we obtained results for the distribution of the fluence of the secondary neutrons and the activated radionuclides in the spherical shells.

Keywords: FLUKA simulations, ^{18}O target, borated polyethylene.

1 Introduction

The Institute for Nuclear Research and Nuclear Energy at the Bulgarian Academy of Sciences (INRNE-BAS) has undertaken the task to build a cyclotron center. The facility is going to be dedicated to applied and fundamental research with radiotracers in areas related to life sciences and industry and to production of well-established radioisotopes for the nuclear medicine [1, 2]. The delivered in 2016 TR24 cyclotron, produced by the Advanced Cyclotron Systems, Inc., has variable energy of the proton beam from 15 to 24 MeV and current up to 400 μA (upgradable to 1 mA). Cyclotron with such parameters is suitable for production of large variety of PET and SPECT radioisotopes. Presently, the main part of the conducted research activities at the cyclotron laboratory are dedicated to numerical studies on the radiological characterization of the setup and capabilities to produce various medical isotopes.

Currently our effort is directed to the production of ^{18}F , since in Bulgaria, the number of nuclear imaging procedures in oncology which are using fludeoxyglucose (^{18}F) keeps increasing over the last 10 years [3]. The ^{18}F is the product of (p, n) reaction on ^{18}O (enriched H_2O liquid target). The irradiation of the target also produces secondary particles (neutrons and gamma rays). It causes activation of long-lived radioactive nuclei in the bunker walls, which leads to a building up of radioactive waste. As suggested and implemented in the practice [4, 5] in our previous studies [6, 7] we tested the idea of local shielding around the liquid target. It has been proven to be useful in limiting the spatial distribution of the secondary particles and thus lower activation of the facility components, longer lifetime of the facility and safer working environment for the personnel has been achieved.

In this work we tested the effect of adding borated polyethylene layer as a local shielding material. We employed a two-step approach for the study. In our previous study we simulated the irradiation of ^{18}O target and obtained collision tape files with secondary neutrons [6]. Here we developed a model with simplified spherical geometry. The source of secondary neutrons (our previously obtained collision files with neutrons) is positioned in the center of the modelling domain. The approach we are employing here is based on Monte-Carlo simulations using computer code FLUKA [8, 9], since it is a well-established tool for target and shielding design, and activation analysis.

2 Description of the Model

The modelling domain for our simulations is shown in Figure 1. In order to make the further descriptions clearer, the simulated geometry is presented, as a three-dimensional “melon slice” through the geometrical center. The previously obtained neutron source [6] is positioned in the center of the geometry. The source of secondary neutrons is obtained through simulation of ^{18}O target irradiation. In the simulation, the emitted neutrons are scored and written in files which are used in this work as a neutron source irradiating the vault. The particles are read one by one from the files and moved to a position defined for simplicity as the center of the spherical geometry. The FLUKA code is transporting the particles, and is doing numerical estimations. In this work we considered a simple scenario – target and respectively local shielding close to the vault walls. In the center of the geometry we have an air-filled sphere with radius of 20 cm containing the neutron source. The air-filled volume is surrounded by

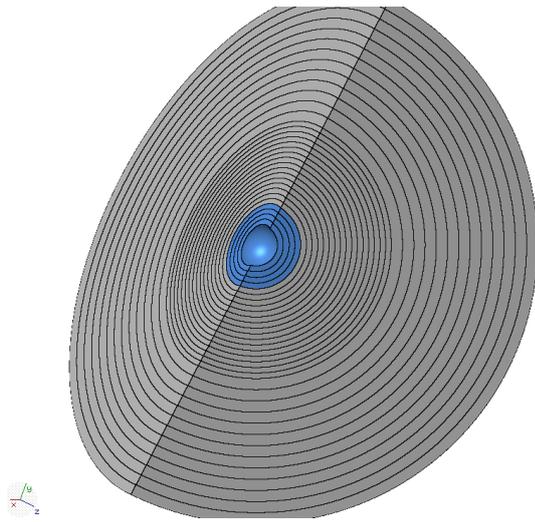


Figure 1. Modelling domain – schematic presentation of the spherical shell and its division into layers (230 cm thick and it is divided into layers – the first 120 cm in layers of 5 cm and the rest into layers of 10 cm). The neutron source is positioned in the geometrical center.

a spherical shell. It is 230 cm thick and it is divided into layers – the first 120 cm in layers of 5 cm and the rest into layers of 10 cm. The main material used to fill the spherical shell is standard concrete with Portland cement (CPC). Its material composition is as follows: 1% H, 0.1% C, 52.9% O, 1.6% Na, 0.2% Mg, 3.4% Al, 33.7% Si, 1.3% K, 4.4% Ca, 1.4% Fe, and its density is $\rho = 2.3 \text{ g/cm}^3$ [10]. The local shielding material that we considered in this study is borated polyethylene (BP) – 12.5% H, 10% B, 77.5% C, with density of 1.0 g/cm^3 [10]. It is well known and widely used material for neutron shielding since boron has good attenuating and absorbing properties for neutron radiation. In our paper are considered three cases: the whole spherical shell is filled with CPC (no local shielding); first 10 cm of the innermost part of the spherical shell filled with BP, the rest is filled with CPC; and the third case – local BP shielding is 20 cm thick.

3 Results and Discussion

For all of the three cases, the obtained specific activities of the generated radionuclides in [Bq/g] are correspondingly shown in the second, third and the fourth column of Table 1. The local exemption limits (LEL), found in “Regulation on radiation protection” [11], for the radioactive materials with weight above 1000 kg are shown in the fifth column. In red are marked the nuclides exceeding the LEL. For the case of CPC filled spherical shielding, we are considering the specific activities in the innermost 5 cm layer. In the other two cases (local shielding of 10 and 20 cm BP) the calculated activities are in the first 5 cm layer of CPC after the local shielding. As it is expected the additional BP shielding reduces considerably the specific activity of the generated radionuclides. The 20 cm BP lead to the reduction in the specific activity of ^{22}Na and ^{55}Fe with one and two orders of magnitude respectively. The isotopes with longer half-life, which exceed the LEL are ^{55}Fe , ^{54}Mn , ^{45}Ca and ^{22}Na . In the case of CPC, they exceed the LEL to

Table 1. Specific activities in [Bq/g] of the nuclides generated in the 5 cm layer, or in the first layer of same thickness just behind the BP shielding (for the two cases – 10 and 20 cm)

| Iso- tope | CPC | 10 cm BP Bq/g | 20 cm BP | LEL | Half-life |
|------------------|------------|------------------|------------|------|----------------------------|
| ^{59}Fe | 11005.6522 | 550.869565 | 72.2608696 | 1 | 44.5d |
| ^{55}Fe | 32435.6522 | 1605.21739 | 264.782609 | 1000 | 2.75a |
| ^{54}Mn | 4699.56522 | 701.73913 | 146.086957 | 0.1 | 312d |
| ^{53}Mn | 6.2426E-05 | 1.4357E-05 | 3.2326E-06 | 100 | $3.74 \times 10^6\text{a}$ |
| ^{51}Cr | 723.913043 | 133.565217 | 31.7956522 | 100 | 27.7d |
| ^{47}Sc | 217.695652 | 27.2043478 | 7.47826087 | 100 | 3.35d |
| ^{47}Ca | 68.8695652 | 8.60869565 | 2.36521739 | 10 | 4.54d |
| ^{45}Ca | 103991.304 | 4608.26087 | 711.304348 | 100 | 163d |
| ^{41}Ca | 11.2869565 | 0.50652174 | 0.07782609 | – | $1.0 \times 10^5\text{a}$ |
| ^{43}K | 1.0054E-06 | 9.3665E-08 | 2.9237E-08 | 10 | 22.2h |
| ^{42}K | 8.6839E-11 | 3.9987E-12 | 6.1183E-13 | 100 | 12.4h |
| ^{40}K | 0.00169413 | 0.00011099 | 1.9581E-05 | – | $1.25 \times 10^9\text{a}$ |
| ^{39}Ar | 403.391304 | 59.826087 | 11.3565217 | – | 269a |
| ^{37}Ar | 840260.87 | 129126.087 | 26137.3913 | – | 35d |
| ^{36}Cl | 0.18013043 | 0.02799522 | 0.00583043 | 1 | $3.02 \times 10^5\text{a}$ |
| ^{26}Al | 0.00048248 | 0.00010274 | 3.112E-05 | 10 | $7.17 \times 10^5\text{a}$ |
| ^{24}Na | 7.6665E-07 | 3.6406E-08 | 5.8422E-09 | 1 | 15h |
| ^{22}Na | 186.826087 | 45.7391304 | 11.6391304 | 0.1 | 2.60a |
| ^{14}C | 0.09386957 | 0.00035765 | – | 1 | $5.7 \times 10^3\text{a}$ |
| ^3H | 8.31304348 | 2.38 | 0.61913043 | 100 | 12.3a |

the following depths: ^{55}Fe – 50 cm; ^{54}Mn – 100 cm; ^{45}Ca – 80 cm; ^{22}Na – 75 cm. For the second considered case (10 cm layer of BP) the respective results are the following: ^{55}Fe – 25 cm; ^{54}Mn – 85 cm; ^{45}Ca – 65 cm; ^{22}Na – 55 cm. And in the case of 20 cm layer of BP: ^{55}Fe – 0 cm (20 cm of BP is enough to reduce its activity below the LEL); ^{54}Mn – 75 cm; ^{45}Ca – 50 cm; ^{22}Na – 40 cm. In Table 2 are shown the specific activities, of the generated radionuclides, in the first 5 cm layer of BP. In this material the only nuclide that exceeds the LEL [11] is ^7Be . It should be noted that in this table the considered LEL is with regards to radioactive materials with weight lower than 1000 kg.

Table 2. Activities – in the BP layer

| Isotopes | BP Bq/g | LEL | Half-life |
|------------------|------------|----------|-----------|
| ^{14}C | 1.27E-02 | 1.00E+04 | 5.7E+03a |
| ^{10}Be | 0.1315 | 1.00E+04 | 1.39E+06 |
| ^7Be | 2246 | 1.00E+03 | 53.2d |
| ^3H | 7.14E+04 | 1.00E+06 | 12.3a |

The results for the distribution in depth of the generated radionuclides are confirmed by the distribution of the neutron fluence shown in Figure 2. As it is expected local shielding of 20 cm BP (Figure 2(b)) limits the penetration of neutrons in the CPC better than that of 10 cm of BP (Figure 2(c)). There is a noticeable difference between the case where we have 20 cm layer of BP (Figure 2(b)) and the one without local shielding (Figure 2(a)).

In Figure 3 the results for the attenuation of the neutron fluence (energy-integrated fluence) with respect to the depth for the three cases are presented. It shows that adding a 20 cm layer of BP reduces on average 4 times the neutron fluence.

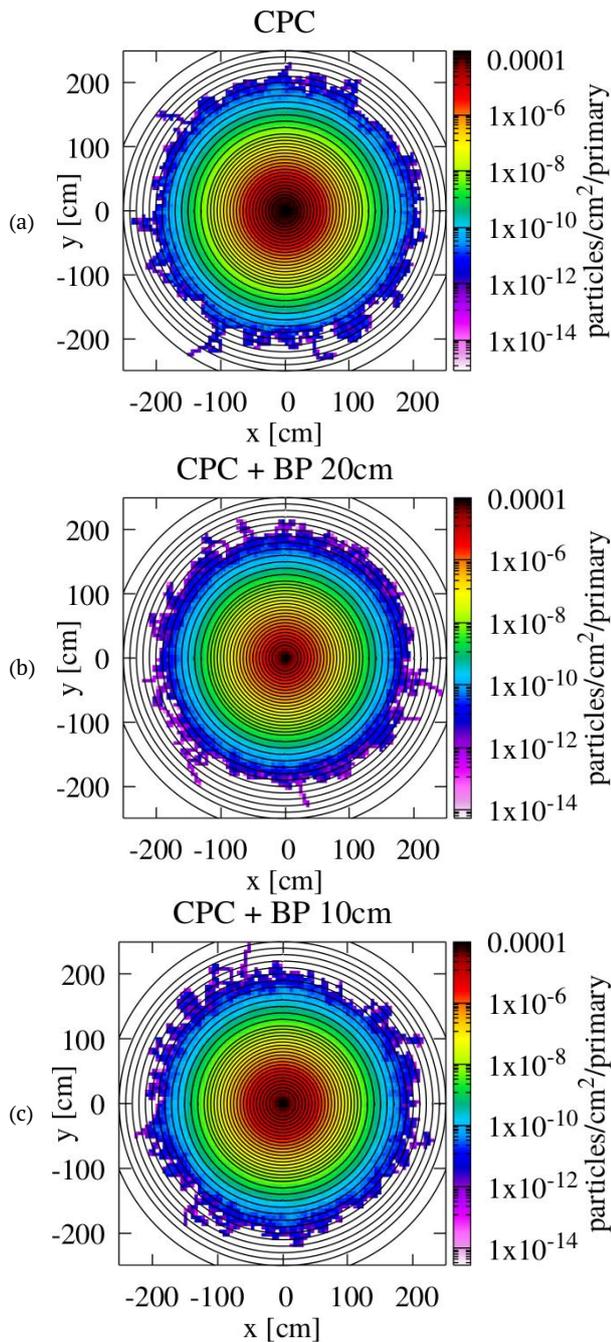


Figure 2. Neutron fluence superimposed on the geometry for the three cases: (a) without; (b) with local target shielding of borated polyethylene – 20 cm; (c) and 10 cm.

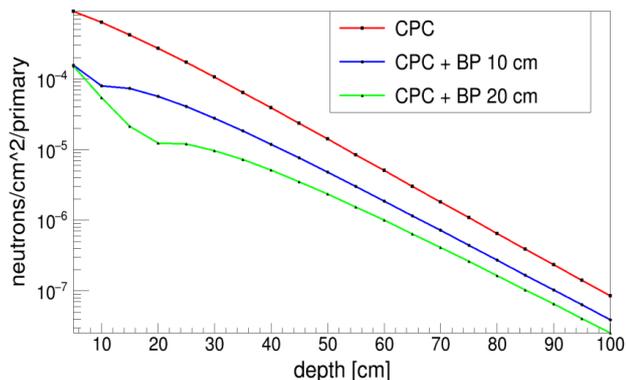


Figure 3. Energy-integrated neutron fluence in relation to the wall depth for the three cases.

4 Conclusion

Evaluation of the distribution of the generated radionuclides and the neutron fluence, for the case of production of ^{18}F is calculated for three cases, using Monte-Carlo simulations. Our results show that adding a layer of 20 cm borated polyethylene around the target reduces considerably the activity of the generated radionuclides and the neutron fluence.

Acknowledgements

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