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# Assessment of the production of medical isotopes using the Monte Carlo code FLUKA: Simulations against experimental measurements



BEAM INTERACTIONS WITH MATERIALS AND ATOMS

Angelo Infantino<sup>a,\*</sup>, Elisabeth Oehlke<sup>b,c</sup>, Domiziano Mostacci<sup>a</sup>, Paul Schaffer<sup>b</sup>, Michael Trinczek<sup>b</sup>, Cornelia Hoehr<sup>b</sup>

<sup>a</sup> Department of Industrial Engineering, Montecuccolino Laboratory, University of Bologna, Via dei Colli 16, 40136 Bologna, Italy

<sup>b</sup> TRIUMF, 4004 Wesbrook Mall, V6T 2A3 Vancouver, BC, Canada

<sup>c</sup> Department of Radiation Science & Technology, Delft University of Technology, Postbus 5, 2600 AA Delft, The Netherlands

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#### 1. Introduction

Cyclotrons are used in nuclear medicine to produce short-lived radionuclides of biomedical interest, especially for positron emission tomography (PET). In the last several years, interest in new isotopes and improved ways of producing established isotopes has driven the need for new target solutions and improved target designs. To mention a few recent works, new production routes are being investigated [1], new designs are being developed for high-power targets [2-4], and liquid targets are being investigated to produce a variety of radiometals [5–9]. In this context, the use of a Monte Carlo simulation for the design and optimization of target holders as well as target materials assumes a very important role in assessing the yield of the desired isotope as well as contaminants inherent to each system, e.g. in the production of <sup>89</sup>Zr [10], <sup>18</sup>F [11], <sup>68</sup>Ga [12], <sup>67</sup>Ga [13], and <sup>68</sup>Ge and <sup>65</sup>Zn [14]. In this work, the Monte Carlo code FLUKA is used. It is a general purpose tool for calculations of particle transport and interaction with matter, covering an extended range of applications spanning from accelerator shielding to target design, calorimetry, activation, dosimetry, detector design, neutrino physics, radiotherapy, and more [15,16]. While the main application of FLUKA is in high energy physics [17], over the past several years FLUKA has also been widely

### ABSTRACT

The Monte Carlo code FLUKA is used to simulate the production of a number of positron emitting radionuclides, <sup>18</sup>F, <sup>13</sup>N, <sup>94</sup>Tc, <sup>44</sup>Sc, <sup>68</sup>Ga, <sup>86</sup>Y, <sup>89</sup>Zr, <sup>52</sup>Mn, <sup>61</sup>Cu and <sup>55</sup>Co, on a small medical cyclotron with a proton beam energy of 13 MeV. Experimental data collected at the TR13 cyclotron at TRIUMF agree within a factor of  $0.6 \pm 0.4$  with the directly simulated data, except for the production of <sup>55</sup>Co, where the simulation underestimates the experiment by a factor of  $3.4 \pm 0.4$ . The experimental data also agree within a factor of  $0.8 \pm 0.6$  with the convolution of simulated proton fluence and cross sections from literature. Overall, this confirms the applicability of FLUKA to simulate radionuclide production at 13 MeV proton beam energy.

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employed in the lower energy regime of medical physics applications, more specifically in proton therapy (PT) [18]. In PT, FLUKA can be employed to simulate tissue activation from the proton beam entering the patient and to assess the absorbed dose. PET isotopes such as <sup>11</sup>C, <sup>13</sup>N and <sup>15</sup>O are produced mainly at the end of the beam range with proton beam energies from several hundred MeV down to threshold [19]. The patient can undergo a PET scan during or after treatment. The PET image can be compared to the simulation, and the location and the magnitude of proton beam energy deposition can be deduced. In this way, PET scans and Monte Carlo simulations together play an important role in the dose verification of the treatment. The quality of the comparison depends strongly on the cross sections used in the simulations [20], and on the Monte Carlo code adopted [21]. To help evaluate the nuclear reaction models in FLUKA, we set out to use FLUKA to simulate the following reactions, all performed at TRIUMF, Canada's national laboratory for nuclear and particle physics:  ${}^{18}O(p,n){}^{18}F$ ,  ${}^{nat}O(p,x){}^{13}N$ ,  ${}^{nat}Mo(p,x){}^{94m}Tc$ ,  ${}^{nat}Ca(p,x){}^{44}Sc$ ,  ${}^{nat}Zn(p,x){}^{68}Ga$ ,  ${}^{nat}Sr(p,x){}^{86}Y$ ,  $^{nat}Y(p,x)^{89}Zr$ ,  $^{nat}Cr(p,x)^{52}Mn$ ,  $^{nat}Zn(p,x)^{61}Cu$ , and  $^{nat}Ni(p,x)^{55}Co$ .

#### 2. Materials and methods

#### 2.1. Cyclotron targetry

All experimental data were measured using the TR13 cyclotron, a self-shielded, external ion source cyclotron that accelerates

<sup>\*</sup> Corresponding author. Tel.: +39 051 2087 702; fax: +39 051 2087 747. *E-mail address:* angelo.infantino@unibo.it (A. Infantino).

negative hydrogen ions to 13 MeV. Protons were extracted from the cyclotron via carbon extraction foils, stripping off the two electrons, thereby reversing the electric charge of the ion and bending its trajectory outwards in the magnetic field. Details of the cyclotron are given in [22,23].

The TR13 cyclotron has two extraction ports each with a target selector. On each target selector, four targets can be mounted. The selector is mounted on a bellows and can be moved horizontally and vertically: When a target is selected for irradiation, the bellows moves it into position in the proton beam. In this work the beam collimation system after the beam extraction, as well as the target assembly for both liquid and solid targets, were simulated with FLUKA, see Fig. 1. On both targets, the proton beam is first collimated by a graphite baffle (1), and is further collimated by two collimator rings (2) and (3), made of anodized aluminum, mounted closely together. The proton beam then travels through a conical graphite collimator cut into four pieces (4). All pieces are electrically insulated from each other. The current created by the collimated proton beam is measured and displayed during irradiation. The target is centered on the proton beam by balancing the current on this last four-piece collimator. Two types of targets were modeled: liquid and solid, see Fig. 1a and b. Both targets are mounted on the target plate via an insulator flange (5), made out of anodized aluminum. The proton beam passes through a 32 mm diameter, 25  $\mu$ m thick aluminum foil that separates the vacuum from the target assembly (between 5 and 6). The foil is cooled by a helium jet (8) through the so-called helium window (6).

In the liquid target, the helium is separated from the target material by a foil of HAVAR, 32 mm in diameter and 39  $\mu$ m thick, placed in front of the target chamber (9) and also cooled by the helium jet (8). HAVAR is a metal alloy with high tensile strength composed of 42.5% cobalt, 13.0% nickel, 20.0% chromium, 2.0% molybdenum, 0.2% carbon, 0.04% beryllium, 1.6% manganese, 2.8% tungsten and the balance iron [24]. At the end of the beam propagation, the proton beam enters the liquid target material which is contained in a chamber 12 mm in diameter and 8 mm deep (9). The target body (7) is made out of niobium or aluminum.

In the solid target assembly, the target foil (10), 32 mm in diameter and of variable thickness is mounted instead of the HAVAR foil, and another helium window (11) is mounted after the target foil to provide additional helium cooling from the back side (13). At the end of the beam propagation, a water-cooled aluminum block (12) is mounted, acting as a beam dump as the target foil is too thin to stop the proton beam completely.

#### 2.2. Monte Carlo model

#### 2.2.1. Geometry and target materials

Two different geometries are created to simulate both target configurations. The collimation systems (1–4 in Fig. 1) are the same for both geometries. The models are created using SimpleGeo [25], a 3D solid modeler, and exported to FLUKA, version 2011.2b.6. The input files are edited with Flair [26], the FLUKA user interface, with which it is possible to define regions, to create and assign materials to regions, to modify the transport and physical parameters, and to manage the scores that are the estimators used to predict the expectation value of one or more quantities, as determined by the radiation field and by the material geometry described by the user. In Flair, every command is passed to the input file through a card, the graphical representation of an input line. In Fig. 1a and b, the models of the two different targets are shown.

The target materials are divided into two overarching categories: liquid targets, which include the water solutions of several salts, and solid targets (Table 1). FLUKA uses the natural isotopic abundance for all elements, but it was decided to input the natural isotopic compositions from the literature [27] into the code to know the exact atomic content: all the isotopes of one element are created with several MATERIAL cards, while the final natural element is created using the COMPOUND card with the appropriate isotopic mass fraction. For all the liquid targets, the mass fraction of all the components of the target mixture is calculated and inserted into FLUKA for the definition of the target material. The main features of the target materials used in this work are reported in Table 1.

#### 2.2.2. Proton beam

Experimental results are compared to simulation with an ideal pencil beam as well as with a realistic beam with a spread in energy and divergence. As a high level of detail is reached in the geometrical model, an accurate model of the source term – the proton beam – is deemed necessary to describe the spread-out beam. In FLUKA, the BEAM card can easily model a particle beam; this



Fig. 1. Cut through the liquid target assembly (a) and of the solid target assembly (b) in the FLUKA Monte Carlo code.

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