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Validating production of PET radionuclides in solid and liquid targets: Comparing Geant4 predictions with FLUKA and measurements

T. Amin[a], A. Infantino[b], R. Barlow[a], C. Hoehr[c,*]

[a] University of Huddersfield, Huddersfield HD1 3DH, UK
[b] European Organization for Nuclear Research, CH-1211 Geneva 23, Switzerland
[c] TRIUMF, Vancouver BC V6T 2A3, Canada

HIGHLIGHTS

- Geant4 useful tool for simulation of PET isotope production.
- Geant4 and FLUKA results consistent.
- Geant4 using TENDL cross sections with QGSP-AllHP model best compromise.
- Model QGSP-BERT-HP and QGSP-BIC-HP do not produce all isotopes.

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ABSTRACT

The Monte Carlo toolkit Geant4 is used to simulate the production of a number of positron emitting radionuclides: ^{13}N, ^{18}F, ^{44}Sc, ^{52}Mn, ^{55}Co ^{61}Cu, ^{68}Ga, ^{86}Y, ^{89}Zr and ^{94}Tc, which have been produced using a 13 MeV medical cyclotron. The results are compared to previous simulations with the Monte Carlo code FLUKA and experimental measurements. The comparison shows variable degrees of agreement for different isotopes. The mean absolute deviation of Monte Carlo results from experiments was 1.4 ± 1.6 for FLUKA and 0.7 ± 0.5 for Geant4 using TENDL cross sections with QGSP-BIC-AllHP physics. Both agree well within the large error, which is due to the uncertainties present in both experimentally determined and theoretical reaction cross sections. Overall, Geant4 has been confirmed as a tool to simulate radionuclide production at low proton energy.

1. Introduction

Radioisotopes play a crucial role in the diagnosis and treatment of cancer. Numerous isotope-producing nuclear reactors are due to end their operation within a few years. As a result, proton-induced reactions have attracted significant interest from the scientific community after cyclotrons proved to be a feasible alternative to reactor produced radioisotopes (Bénard et al., 2014; Schaffer et al., 2015). Currently cyclotrons can be used to produce radioisotopes for imaging techniques such as positron emission tomography (PET) and single photon emission computed tomography (SPECT). The irradiated target can be in solid, liquid or gaseous form and may be required to satisfy strict design constraints. For example, a target may have material composition restrictions to achieve a desired specific activity, proton energy constraints to avoid unwanted isotope production, or may need to survive several hours of proton irradiation without any thermal issues. As a result, cyclotron targets and materials can be very expensive. Monte Carlo (MC) simulations can be used to assess the expected yield and to optimize target design and materials to maximize yield of the isotope of interest without increasing the production of contaminants (Infantino et al., 2011; Remetti et al., 2011; Sadeghi et al., 2013; Fassbender et al., 2007). The success in using MC for yield assessment depends strongly on the cross section data used for the simulation. Despite a large number of experiments carried out with proton activation, the data available are often inconsistent and at times data from different experiments conflict with each other.

In this work, the MC package Geant4 has been used to simulate the yields of the following PET isotopes: ^{13}N, ^{18}F, ^{44}Sc, ^{52}Mn, ^{55}Co ^{61}Cu, ^{68}Ga, ^{86}Y, ^{89}Zr and ^{94}Tc. The results have been compared to our previous work with the MC package FLUKA and with experiments (Infantino et al., 2016). Different physics models in Geant4 have been tested to find the best approximator of isotopic yield to experiments. Previous results for ^{13}N, ^{18}F, and ^{68}Ga have been published (Amin et al., 2017) and are repeated here for completeness.

* Corresponding author.
E-mail address: choehr@triumf.ca (C. Hoehr).

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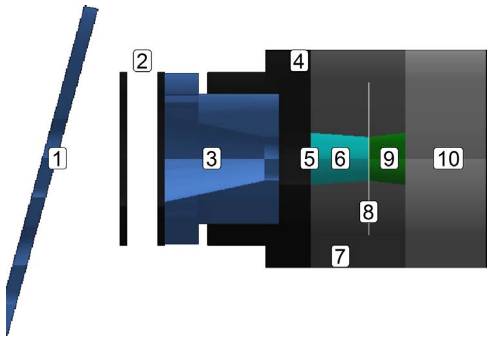

Fig. 1. Liquid target.

2. Materials and methods

2.1. Experiments

The experimental details have been described and, where appropriate, referenced by Infantino et al. (2016). Some details are repeated here for the convenience of the reader.

The TR13 cyclotron is located at TRIUMF, Vancouver, Canada and used for routine production of medical isotopes. It is self shielded and accelerates negative hydrogen ions to 13 MeV energy with currents of routinely up to 25 μA. Extraction occurs with the use of a carbon foil which strips off the two electrons thus reversing the charge and bending trajectory of the ion in the magnetic field. The cyclotron has two extraction ports with a target selector, which can move the target into the proton beam. Further details of the cyclotron are provided by Laxdal et al. (1994); Buckley et al. (2000).

The selector has four positions, allowing eight different targets to be installed at a time. Two target assemblies were simulated in Geant4. Figs. 1 and 2 illustrate the liquid and solid target assemblies respectively with each component labeled numerically. The proton beam enters the assembly though the baffle (1) and collimator rings (2). The beam is then collimated further with a four quadrant conical collimator (3) contained within an insulator flange (4). Each quadrant of the collimator is capable of measuring beam current separately and the four readings can be used to deduce the position of the proton beam. The beam then enters the target assembly through a 25 μm thick aluminium foil (5), which separates the cyclotron vacuum from the target assembly. Due to the power deposition in the foil, helium cooling (6) is applied to the foil in the helium window (7).

The liquid target (9) is a closed volume of 0.9 ml capacity, with 8 mm depth and 12 mm diameter. The liquid target is separated from the helium cooling (6) by a HAVAR foil (8). HAVAR is a cobalt based metal alloy with high tensile strength. It is 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 remainder iron, see Hamilton Precision Metals (2017). The target body (10) is composed of standard niobium. Target loading and unloading is performed using an automated loading system, see Hoehr et al. (2014).

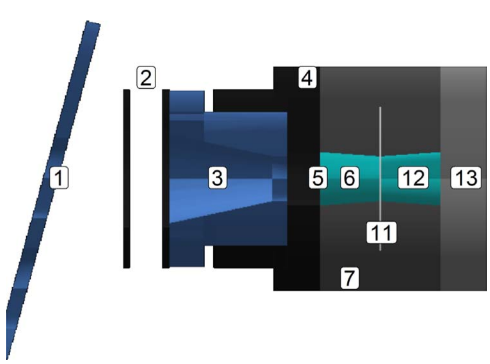

Fig. 2. Solid target.

In the solid target assembly, the foil target (11) is in the place of the HAVAR with helium jets for cooling on both sides (6) and (12). Due to the use of thin foils, the proton beam traverses through the target and is finally stopped by the water cooled aluminium block (13) which acts as the beam dump. The geometries were modelled as accurately as possible by using dimensions from technical drawings.

The nuclear and chemical properties of the liquid and solid target materials are listed in Table 1. After irradiation, isotopic yield measurements were performed using gamma-ray spectrometry analysis or ionization chamber measurements. All measured yields were decay-corrected to the end of bombardment (EOB). When multiple irradiations took place for the same isotope, the yield was normalized to the beam current prior to calculating the average saturation yield. The error in the yield is dominated by the standard deviation of the different irradiations. For more details see Infantino et al. (2016) and references therein.

2.2. Monte Carlo simulations

Geant4 is an all particle Monte Carlo toolkit designed for simulating particle interactions from 100 TeV down to a few eV. Geant4 is implemented in C + + and has great flexibility and expandability and thus is used in various applications such as space research, Large Hadron Collider (LHC) experiments, medical physics or microdosimetry applications (Agostinelli et al., 2003; Allison et al., 2006, 2016).

2.2.1. Saturation yields

In Geant4, the calculation of induced activity relies on the cross section library used for the inelastic nuclear reactions. These cross sections are included in the TENDL1.3 package and can directly calculate the number of isotopes produced. The production rate for each isotope is simulated taking into account primary proton impact, secondary interactions and decay of other isotopes produced in these interactions. Geant4 calculates the isotope production from the primary particle induced production and also the full Bateman solution considering the breeding of radioactive decay products. The number of isotopes at any given time t during the irradiation is given by:

$$N(t) = \frac{nI}{n_p e\lambda}[1 - e^{(-\lambda t)}] \quad (1)$$

where n is the number of isotopes produced per unit mass and unit time (a function of the proton flux, the target density, and the nuclear cross section), I is the proton beam current from the accelerator, n_p is the number of incident protons, e is the proton electric charge, and λ is the decay constant of the isotope, see Bungau et al. (2014). This equation reaches a saturation level for long irradiation, N_{sat}, where $N_{sat} = \frac{nI}{n_p e\lambda}$. Using $A_{sat} = N_{sat}\lambda$, the saturation yield Y_{sat} in $Bq/\mu A$ is given by:

$$Y_{sat} = \frac{A_{sat}}{I} \quad (2)$$

When calculating yield ratios the experimental and MC uncertainties have been added in quadrature.

2.2.2. Target geometry and material definition

The solid and liquid targets have been represented in Geant4 using two geometries as shown in Figs. 1 and 2. Geant4 provides a wide range of simple solid geometries that can be used. More complex geometries such as the conical collimator can be generated by combining existing shapes with Boolean operators such as G4UnionSolid and G4SubtractionSolid.

The target materials have been divided into two categories: liquid target, containing water solution of salts, and solid targets. While it is possible to use the natural isotopic composition of elements from the NIST database, user defined isotopic compositions were used in order to