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Monte Carlo modeling provides accurate calibration factors for radionuclide activity meters

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HIGHLIGHTS

- We developed a Monte Carlo model of a radionuclide activity meter using Geant4.
- The model was validated using several reference sources and experimental trials.
- Validation provided very accurate results (less than 4% discrepancy).
- This paper shows how simulations establish a tool for many applications.
- Examples are reported, such as evaluation of new or optimized calibration factors.

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ABSTRACT

Accurate determination of calibration factors for radionuclide activity meters is crucial for quantitative studies and in the optimization step of radiation protection, as these detectors are widespread in radiopharmacy and nuclear medicine facilities. In this work we developed the Monte Carlo model of a widely used activity meter, using the Geant4 simulation toolkit. More precisely the "PENELOPE" EM physics models were employed. The model was validated by means of several certified sources, traceable to primary activity standards, and other sources locally standardized with spectrometry measurements, plus other experimental tests. Great care was taken in order to accurately reproduce the geometrical details of the gas chamber and the activity sources, each of which is different in shape and enclosed in a unique container. Both relative calibration factors and ionization current obtained with simulations were compared against experimental measurements; further tests were carried out, such as the comparison of the relative response of the chamber for a source placed at different positions.

The results showed a satisfactory level of accuracy in the energy range of interest, with the discrepancies lower than 4% for all the tested parameters. This shows that an accurate Monte Carlo modeling of this type of detector is feasible using the low-energy physics models embedded in Geant4. The obtained Monte Carlo model establishes a powerful tool for first instance determination of new calibration factors for non-standard radionuclides, for custom containers, when a reference source is not available. Moreover, the model provides an experimental setup for further research and optimization with regards to materials and geometrical details of the measuring setup, such as the ionization chamber itself or the containers configuration.

1. Introduction

Radionuclide activity meters (frequently, even if improperly, termed as "dose calibrators") are re-entrant ionization chamber detectors which are primarily used to assay the amount of radioactivity contained in radiopharmaceutical preparations, and are thus

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widespread in radiopharmacies and nuclear medicine facilities. These devices are designed to measure the charge produced within the internal gas through interaction with the ionizing radiation (mostly gamma photons) emitted from the sample placed inside the well. Several methods have been established (Zimmerman and Cessna, 1999) to determine the calibration factors, which must be applied in order to convert the reading of the equipment in terms of current into an activity measurement. Among these we mention: the use of sources containing a certified amount of activity of the radionuclides of interest (IAEA, 2006); the use of a long-lived certified source which has similar emission as that of a short-lived nuclide (Zimmerman and Cessna, 2010) and cross-calibrated with the radionuclide of interest ("mock" sources); the inter-comparison against an independently calibrated instrument (Capogni et al., 2008).

However, at least in some cases (i.e. radionuclides of infrequent use or assessment of the contribution of radionuclidic impurities), calibration factors continue to be derived from the best fit curve of known energy-response points by knowing the energies and relative intensities of each gamma emission of the nuclide of interest (NCRP, 1985).

Activity meter calibration procedures are critical to obtain accurate values for the activity of radiopharmaceuticals administered to patients in routine Nuclear Medicine applications, which is fundamental for quantitative studies and a necessary component of the optimization of radiation protection. The geometry of the container (shape and material, filling and position) must also be considered when performing a measurement, and these elements are usually more significant when the source emits beta particles or high intensity, low-energy photons.

The aim of this work was to develop and validate a Monte Carlo model for a widely used activity calibrator, using Geant4. Primarily, the model can in fact provide an independent tool for the computation of the calibration factors, as well as the correction factors needed for different container geometries. This is of great advantage for the measure of novel or non-standard radiopharmaceuticals (in particular, in the case of very short-lived nuclides), for which a calibrated reference source may not be readily available and/or only small amounts of radionuclide are available for experimental tests.

Moreover, the Geant4 model of the detector, once validated, establishes a convenient setup for studying the response of the chamber under different conditions, since it allows a highly customizable particle tracking and scoring. Plus, the latest versions of the Geant4 package, which remains a widespread tool in high energy physics experiments, are seeing increased popularity in the field of medical physics, as the low-energy EM interaction models are now validated and are accurate enough for simulations of detectors such as the one object of this work.

2. Materials and methods

2.1. The activity meter

We modeled the activity meter "MP-DC", manufactured and distributed by MecMurphil srl (Ferrara, Italy) under Murphil srl (www.murphil.eu) license and installed in a research laboratory at our institution. The previous version of MP-DC activity meter, known with the registered brand name of TALETE/PITAGORA, has been installed worldwide in hundreds of nuclear medicine and PET facilities. Beside the classical design of re-entrant ionization gas chambers, it features a newly-designed electrometer and application oriented software. The internal sensitive volume is 28 cm height and filled with pure argon at 15 bar pressure, contained in aluminum cylindrical walls. The internal wall of the chamber is 70 mm in diameter and 1.5 mm thick. The device at our disposal is the stand-alone version of the chamber, which is suitable also for use outside of manipulation cells and it is shielded from external radiation with a 6 mm thickness of lead. The nominal potential between the anode and cathode is 500 V. The electrometer board, based on digital components, features proprietary firmware which minimizes the measurement errors through a dynamic adaptation of the integration time of the ionizing current. The measurement of the collected charge is performed by the operator from a separate console which essentially converts the charge collected per unit time, I, to the activity of the sample A using a calibration factor K_{abs}:

$$A = K_{abs} \cdot I \tag{1}$$

2.2. Monte Carlo model

Geometry of the calibrator and particle physics were modeled using the Monte Carlo simulation toolkit Geant4 version 9.4 (Agostinelli et al., 2003; Allison et al., 2006). More precisely, the low-energy physics package "PENELOPE" was used. The full decay of radionuclides of interest was included in the simulations, considering transport and interaction of photons and electrons, including all effects, such as Compton scattering, photoelectric absorption, electron and positron scattering and ionization, pair production and bremsstrahlung radiation. In contrast to the standard EM (Electromagnetic cascade emission) package, which is normally provided as a default for Geant4 simulations, this list allows for simulations down to lower energies (250 eV) (Chauvie et al., 2004). Moreover the low-energy models include atomic processes, such as X-ray fluorescence and Auger electrons emission, and are based on evaluated libraries EPDL97, EEDL and EADL (Evaluated Atomic Data Library) (Guatelli et al., 2007; Ivanchenko et al., 2011). In this work, a range cut-off of 10 μm has been set for secondary particles production, which implies the simulation down to 250 eV in gaseous materials for both gamma and beta particles.

The geometry of the chamber was carefully reproduced using a detailed schematic provided by the factory and according to the tolerances of the computer controlled machining. As shown in Fig. 1, elements such as electrode, guard ring and shielding were included in the model, as was the sample holder. Several components were critical for the correct modeling of the response of the chamber, such as the internal wall thickness. The correct definition of materials of the constituents was also very important. While for the simplest elements and materials we relied on the NIST database, which is integrated in the Geant4 libraries, some compound materials were defined using the fractional mass of each component element. This was the case, for example, of the aluminum alloy which forms the walls of the chamber, which contains small amounts of iron and magnesium. The filling gas has been modeled according to the manufacturer specifications (argon, 15 bar).

The position, shape, energy and type of sources were selected using the Geant4 module *G4SingleParticleSource*. While gamma and beta particles may be directly generated, this class also allows for simulation of unstable nuclei. In this way, for each event the radionuclide of interest was generated at rest, and the Geant4 code tracked it as an unstable particle so that the full decay scheme was reproduced, i.e. as the decay channel was sampled for each event, the correct relative intensities of gamma and beta emissions were observed. The radioactive decay model in Geant4 is empirical and data-driven, and uses the Evaluated Nuclear Structure Data File (ENSDF) (Tuli, 1987) for information on branching ratios, nuclear level structure and energy of the decay process. This mode was chosen for most of the simulations in this work, as complex decay schemes could be easily and accurately reproduced.

In order to assess the response, or sensitivity, of the modeled activity meter, we assumed the ion chamber to operate in