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A radionuclide calibrator based on Cherenkov counting for activity measurements of high-energy pure β^--emitters

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ABSTRACT

Due to their stability and reproducibility, re-entrant pressurized ionization chambers (also called radionuclide calibrators) are widely used for activity measurements in nuclear medicine services as well as in national metrology institutes to maintain reference standards. Generally, these secondary instruments yield accurate activity measurements for γ-emitting radionuclides. Ionization chambers are easy to use and thus well-adapted to guarantee the metrological traceability between national metrology institutes and end-users. However, the reproducibility of calibration factors can be significantly impaired when measuring high-energy pure β^--emitters such as radiopharmaceuticals based on ^{90}Y. This is because the bremsstrahlung emission contributing to the instrument response is strongly dependent on the geometry of the components surrounding the radioactive solution. The present article describes a new design based on pulse counting to address this problem. It takes advantage of Cherenkov emission resulting from Compton scattering in transparent materials. The interest of Cherenkov counting is to obtain a low-sensitivity detector that enables direct measurements of high-activity solutions (at least up to 10 GBq for ^{90}Y-microspheres in aqueous suspensions used in nuclear medicine). A simple design based on a geometry close to an ionization chamber used at LNHB (Vinten 671 type) is described. The feasibility in terms of detection efficiencies (lower than 10^{-4} for ^{90}Y solutions) of the new radionuclide calibrator is investigated by Monte Carlo calculations using the Geant4 code.

1. Introduction

In the field of radionuclide metrology, re-entrant pressurized ionization chambers (also called radionuclide calibrators) are secondary instruments commonly used in national metrology institutes (NMIs) to maintain and to transfer reference standards of activity. These devices are also widely applied in nuclear medicine services for activity measurements of radiopharmaceuticals dedicated to diagnosis or therapeutic applications. Due to their stability and reproducibility, ionization chambers (ICs) represent a useful detection system to guarantee the metrological traceability of end-users' measurements to national activity standards (Zimmerman and Judge, 2007). At the international level, the Bureau des Poids et Mesures (BIPM) has in charge the equivalence of activity standards between NMIs through international comparisons. To this end, the Système International de Référence relies also on the use of a re-entrant IC since 1976 (Michotte, 2002; Ratel, 2007).

The IC principle remains unchanged since several decades (Schrader, 2007). The classical design consists in a gas-filled cylindrical vessel with a well at the centre of which containers (ampoules, vials, syringes, etc.) filled with radioactive solutions are placed to obtain measurements in quasi 4π geometry. The activity is determined by measuring an ionization current proportional to the energy deposition rate of ionizing radiation in the gas medium. For that purpose, calibration factors needs to be determined using radionuclide solutions traceable to primary standards in NMIs.

ICs are mostly sensitive to x or γ photon emission following radionuclide disintegrations. Thus, calibration factors with low uncertainties (less than 1%) and good reproducibility are currently obtained in NMIs for activity measurements of γ-emitters. But in the case of high-energy pure β^--emitters such as ^{90}Y (E_{max} ~2280 keV), the response of ICs predominantly relies on the detection of bremsstrahlung emitted along the track of beta particles in the container and its surroundings (e.g. source holder). The reproducibility of the measurements is then impaired by their higher sensitivity to variations in the geometrical characteristics of those components, e.g. container wall thickness.

In order to diminish the impact of such problems, the approach described in the present article is to implement pulse counting instead of measuring an ionization current. But then the possibility of counting

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saturation due to the high-activity of radioactive solutions must then be prevented (e.g. up to 10 GBq for ^{90}Y-microspheres in aqueous suspensions such as SIR-Spheres® (Sirtex, Australia)). In addition, it is intended to preserve most of the practical features of ICs: user-friendly instrument, use of the same containers for radioactive solutions (ampoules, vials, syringes, etc.), quasi 4π geometry. The solution presented here is to use a low-sensitivity detector based on Cherenkov light emission.

The paper first recalls the main characteristics of typical ICs applied as secondary instruments and the related problems when measuring high-energy pure β^{-}-emitters such as radiopharmaceuticals based on ^{90}Y. A description of the new design based on Cherenkov counting and the related advantages are exposed afterwards. Monte Carlo calculations using the Geant4 code (Agostinelli et al., 2003) are finally presented to give a first validation of the capability of the new radionuclide calibrator to measure the activity of ^{90}Y solutions up to 10 GBq with an appropriate low-detection efficiency.

2. Design of a new radionuclide calibrator based on Cherenkov counting

2.1. Problems related to activity measurements of high-energy pure β^{-}-emitters with ionization chambers

Typical re-entrant ICs (or radionuclide calibrators) operating in hospitals and in NMIs for activity measurements can be described as gas-filled vessels constructed with coaxial cylinders and equipped with a well at the centre of which radioactive solutions are placed. The dimensions of the detection system are generally chosen to obtain a quasi 4π detection geometry. For γ-emitting radionuclides, the mechanism of energy deposition leading to gas ionization is mostly due to the production of secondary electrons through Compton and photoelectric interactions in the gas and other elements (walls and electrodes). The voltage applied between the electrodes is set to reach a saturation current that minimizes the recombination of ion pairs and the effect of possible voltage fluctuations. As a result, the measured current is proportional to the radionuclide activity and the related calibration factor is determined by means of solutions of known activities of the same radionuclide, traceable to primary standards. These parameters are specific to the radionuclide calibrator used, and for a given instrument, to the geometry of the source. Accurate and reproducible activity measurements are usually achieved for γ-emitting radionuclides (associated uncertainties lower than 1%).

In the case of high-energy pure β^{-}-emitters, the ionization current results predominantly from the interaction of bremsstrahlung produced inside the container and its surroundings (e.g. source holder). The probability of direct interaction of beta particles in the gas medium is relatively low (depending on their energy). The variability of the energy deposition in the gas medium is thus mostly related the possible variation of the parameters that influence the emission rate of bremsstrahlung photons and their energy distribution.

This issue has been largely discussed in the literature (Coursey et al., 1993; Santos et al., 2011; Fenwick et al., 2014). Recently, such a problem of reproducibility was observed for the IC calibration of ^{90}Y-labelled resin microspheres SIR-Spheres® developed for the treatment of liver tumors by radioembolization (~3 GBq injected to patients). Uncertainties up to 5% were reported on calibration factors due to the non-homogeneity of the vials provided by the manufacturer (Ferreira et al., 2016; Thiam et al., 2016).

2.2. Main characteristics of Cherenkov effect for its application in a radionuclide calibrator

As only a fraction of the deposited energy is needed to exceed a detection threshold, a pulse-mode counting can be implemented in the new transfer instrument to lower the variability of activity measurements for high-energy pure β^{-}-emitters. For instance, the pulse mode is applied at the BIPM for the SIR transfer instrument (SIRTI) to establish a link between the SIR and NMIs (Michotte et al., 2013) for short-lived radiopharmaceuticals (^{99m}Tc, ^{18}F). The SIRTI system is based on a well-type NaI(Tl) scintillation detector which is limited in terms of high-activity measurements due to its high sensitivity.

In order to allow high-activity measurements (up to 10 GBq for ^{90}Y solutions) using such a pulse-mode instrument, it is mandatory to have a low-sensitivity detector. As described in Fig. 1, the basic idea of the new design for a radionuclide calibrator is to replace the gas medium by a transparent dielectric material, and to measure Cherenkov photons which are produced in it by secondary electrons released following the interaction of bremsstrahlung.

Cherenkov light is generated when a charged particle travels through a transparent dielectric medium with a speed greater than the phase velocity of light in that medium. The threshold condition of Cherenkov effect is given by the relation $n\beta > 1$ (where n is the refractive index of the medium; β is the ratio of the velocity of the charged particle to the speed of light in vacuum). Cherenkov photons are emitted from the surface of a cone at an angle θ with respect to the path direction of the charged particle. The angle is defined by the following expression $\cos\theta=(n\beta)^{-1}$.

The minimum energy of electrons E_{th} (in keV) for Cherenkov emission depends on the refractive index of the medium and is obtained from special relativity:

$$E_{th}=511\bullet\left[\left(1-\frac{1}{n^{2}}\right)^{-1/2}-1\right].$$

Refractive indices depend on the materials used as well as on photon wavelengths. In the case of fused silica, n is equal to 1.47 at the wavelength of 400 nm and yields a Cherenkov threshold of about 186 keV; at 200 nm, n is equal to 1.55, thus leading to a lower threshold (158 keV). Higher detection thresholds can be achieved with materials such as silica aerogels (Cantin et al., 1974) having refractive indices lower than 1.1 (corresponding to a Cherenkov threshold greater

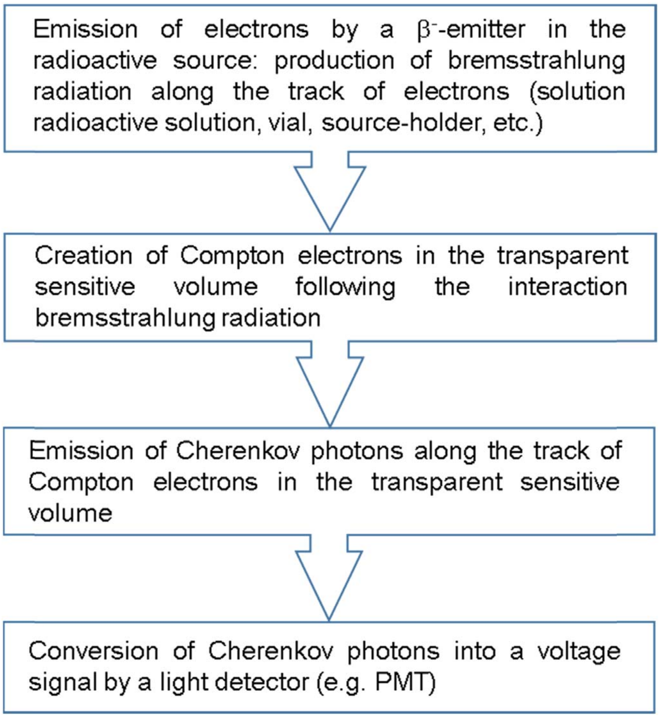

Fig. 1. Diagram describing the different interactions in the new design leading to the production of Cherenkov photons in a transparent material replacing the gas medium in ICs.