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Comparison of experimental and calculated calibration coefficients for a high sensitivity ionization chamber

M.N. Amiot *, M.R. Mesradi, V. Chisté, M. Morin, F. Rigoulay

CEA-LIST/LNE-LNHB, CE Saclay, 91191 Gif sur Yvette Cedex, France

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ABSTRACT

The response of a Vacutec 70129 ionization chamber was calculated using the PENELOPE-2008 Monte Carlo code and compared to experimental data. The filling gas mixture composition and its pressure have been determined using IC simulated response adjustment to experimental results. The Monte Carlo simulation revealed a physical effect in the detector response to photons due to the presence of xenon in the chamber. A very good agreement is found between calculated and experimental calibration coefficients for 17 radionuclides.

1. Introduction

Pressurized ionization chambers (IC) are used in nuclear medicine services and in metrology laboratories for standard activity measurements. The instrument response depends, among the design of the chamber (wall thickness, nature of the filling gas, its pressure, etc.), on the dimensions of the source (volume of syringe, vial, capsule, ampoule, etc.) and on the shielding of the ionization chamber. Calibration coefficients are specific to each kind of radionuclide calibrator, each radionuclide and each type of sample composition and container (Ceccatelli et al., 2007); (Olsovcová and Havelka, 2006; Olsovcová, 2010). Several authors have determined the detection efficiency of different chamber models through mathematical curves (Michotte et al., 2006; Schrader and Svec, 2004; Schrader et al., 2008) or through Monte Carlo simulation. Monte Carlo simulation presents the advantage of making possible the determination of the ionization chamber response for radionuclides contained in any matrix, volume and in any container. The first studies were undertaken with the simulation of the IC response to pure gamma emitters in glass ampoules using the Monte Carlo codes EGS4 (Suzuki et al., 1998; Sato et al., 2006), Geant (Gostely and Laedermann, 2000; Seneviratne et al., 2007), PENELOPE (de Vismes and Amiot, 2003; Amiot, 2004; Kryeziu, 2006; Kryeziu et al., 2007) and finally using the code MCNP (Aleissa, 2002; Olsovcová, 2004). In this work, the response function of a specific highly-pressurized ionization chamber filled with a mixture of gases is studied using the Monte Carlo code PENELOPE. Calibration coefficients were determined using the calculation results obtained with that code and compared with the experimental ones for 17 radionuclides.

2. Experimental method

2.1. Experimental set-up

The Vacutec chamber n° 70129 (Nuklear-Medizintechnik Dresden GmbH, Germany) is a pressurized IC filled with a mixture of xenon and argon gases. The mass percentage of each gas was not previously known but was determined in this study using experimental values. The total pressure given by the manufacturer is 1.1 MPa. The material thicknesses and dimension measurements were performed at LNHB using X-ray radiography presented in Fig. 1. It is a small cylindrical chamber made of aluminum alloy and has a 1 mm wall in the re-entrant well and a 0.5 mm electrode with external dimensions of 26 cm height and 10 cm diameter. A 0.5 mm thick polymethyl methacrylate (PMMA) liner was introduced in the well of the chamber in order to protect against contamination of the inner wall. The chamber is surrounded by a 3 mm thick copper shield which is also surrounded by a 5 cm thick lead shield so that lead fluorescence will not be detected by the chamber as it has already been observed (de Vismes and Amiot, 2003). A 6517 Model Keithley electrometer is used to integrate the IC charges and transfer data via an interface bus (GPIB) and also to supply the IC high voltage (−400 V). A home-made program using Hewlett-Packard HP-Vee® controls the current and background measurements, as well as activity and decay correction calculations. The whole measurement system stability is checked using a ^{226}Ra source. A typical measured sample is a 5 ml aqueous active solution contained in a glass

* Corresponding author. Tel.: +33 169083689; fax: +33 169082619.
E-mail address: marie-noelle.amiot@cea.fr (M.N. Amiot).

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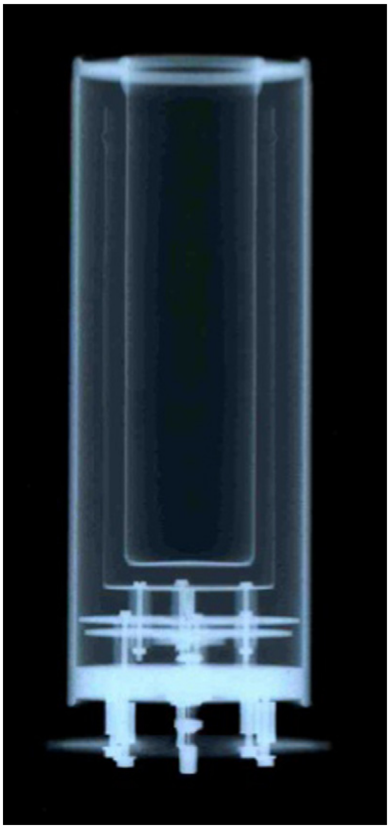

Fig. 1. A LNHB Vacutec® IC radiography.

sealed ampoule (wall thickness: 0.61 ± 0.01 mm). It is positioned on a PMMA stand placed at the bottom of the well of the chamber.

2.2. Ionization chamber calibration

When calibrating an IC, the first step is the provision of standards. At LNHB, our chamber is calibrated with very well defined standards and calibration coefficients were deduced from absolute activity measurements performed by our laboratory within the frame of international comparisons or contributions to the International Reference System (SIR; Rytz, 1983). Most of the active solutions prepared by the laboratory are in a hydrochloric acid medium (0.1 mol/L) with a specific carrier concentration adapted to the chemical properties of the radionuclide. Radionuclide purity tests are systematically undertaken using X- and gamma-ray spectrometry (coaxial HPGe $\approx 100\ cm^3$ and planar HPGe). A minimum of three 5 ml LNHB standard ampoules are prepared in order to check the homogeneity of the active solution. The radionuclides studied are ^{125}I, ^{123}I, ^{67}Ga, ^{111}In, ^{201}Tl, ^{57}Co, ^{56}Co, ^{60}Co, ^{18}F, ^{11}C, $^{99}Tc^m$, ^{85}Sr, ^{75}Se, ^{54}Mn, ^{65}Zn, ^{22}Na, ^{88}Y.

3. Monte Carlo simulation method

The aim of this work is to determine the IC ionization chamber response in order to be able to calculate calibration coefficients for gamma emitting radionuclides. The calculated calibration coefficients are obtained using the total mean energy deposited per emitted photon determined using the Monte Carlo code PENELOPE-2008 (Salvat et al., 2008) as described in (de Vismes and Amiot, 2003) and (Amiot, 2004).

3.1. W value determination

The average energy spent in the production of an electron–ion pair is needed when calculating the calibration factor. Nevertheless, two major difficulties appear in the present experimental set-up used to determine this parameter: the filling gas is a mixture of two gases (argon and xenon) and the pressure is high (around 1 MPa). The first *W*-values measured for argon and xenon mixtures with different amount of gases were published in 2008 (do Carmo et al., 2008). We then used the *W* value equal to (21.63 ± 0.21) eV given in this article for a 60% Ar and 40% Xe mixture which is very close to the mass percentage of the two constituents determined hereafter. It has to be noted that the results presented in this article were obtained for a pressure of 0.1067 MPa whereas the pressure of the filling gas determined in this work is 0.975 MPa. The presence of a pressure dependence of *W* values has been observed for high pressures (Xe pressure above 5 MPa) (Bolotnikov and Ramsey, 1997) and for low pressures in the 7–133 kPa pressure range (Parks et al., 1972). In both articles, a decrease of the *W* value was observed with the increase of the pressure as it was already assumed in the ICRU Report 31 (1979). On the other hand, Borges and Conde (1996) were not able to identify any pressure dependence for pure xenon within the limit of the experimental uncertainty in the 38–125 kPa pressure range. A dependency of the *W* value for intermediate pressures between 0.1 MPa and 5 MPa might exist but there is insufficient data available to make a valued judgment. Therefore no adjustment was made to the *W* value in this work.

3.2. Simulation parameters

PENELOPE simulation parameters chosen under our experimental conditions are the same as those determined in previous studies (de Vismes and Amiot, 2003) and (Amiot, 2004). The simulations were performed using more than 10^6 histories in order to keep the relative statistical uncertainty of the Monte Carlo simulation below 1% for each run. All uncertainty values in the following text are standard uncertainties with a coverage factor $k=1$.

3.2.1. Nuclear data

The nuclear data used were extracted from the Nucléide software (Bé et al., 1999; Dulieu et al., 2004) except for the ^{67}Ga for which the γ-emission intensities used were taken from Bobin et al. (2007).

3.2.2. Ionization chamber modeling

The model of the ionization chamber has been established using the manufacturer's specifications for the type of materials used. The main materials involved are HCl 0.1 M for the solution which contains the radionuclide (and H_2O specifically for ^{125}I, ^{123}I and $^{99}Tc^m$ radionuclides), glass for the 5 ml FIOLAX® ampoules whose composition has been determined (Iroulard, 2007), PMMA, air, PVC, aluminum, 64% of argon and 36% of xenon mass percentages (determined in this work see Section 4), bakelite, copper and lead. Eleven materials were used for the modeling of the chamber. The active volume was approximated by the geometric volume.

3.2.3. Filling gas density calculation

The density of the gaseous mixture under pressure is not easy to calculate. Tournier and El-Genk (2008) give an equation of state which enables the calculation of the compressibility factor for pure gases. This factor is important since its value deviates from unity by several per cent for pure xenon and for pressure around