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Simulation of the response of an ionization chamber to ²¹⁴Bi emission.

Application to the measurement of ²²²Rn.

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Abstract

PENELOPE simulations of a Vinten ionization chamber (IC) were performed to investigate the influence of the thickness of glass-ampoules used in ²²²Rn standardization. The simulation reveals a non-negligible variation of the energy deposited in the chamber gas region (which may induce a proportional variation of the measured current) when considering the β transition emissions of the daughters of ²²²Rn. This reinforces the idea of using a specialist container (made of metal to preserve the integrity of the container) that would circulate between the metrology laboratories in the context of international comparison exercises using the BIPM international reference system (SIR)..

Keywords: Ionization chamber, Monte Carlo simulation, PENELOPE, Radionuclide metrology, Radon.

1. Introduction

Well-type ionization chambers (ICs), connected to current-measuring electronics, are activity measurement instruments commonly used in nuclear medicine services, national metrology institutes (NMI) or radiopharmaceutical industry. It is also the transfer instrument used for the Système International de Référence (SIR) for activity comparison at the Bureau International des Poids et Mesures (BIPM) (Rytz, 1983, Ratel, 2007). The accuracy of the results and the ease of measurement explain the choice of this technique. The response of such instruments *i.e.*, the measured ionization current (which is related to the total energy deposited in the IC gas volume), depends on the chamber design (wall thickness, nature and pressure of the filling gas), the nature and dimensions of the source (type of vial or ampoule, filling height of radioactive sample, etc.), and the shielding around (Ceccatelli et al., 2007; Kryeziu et al., 2007; Thiam et al., 2016). Generally, ICs are well adapted for solutions of gamma-emitting radionuclides (Amiot et al., 2012; Zimmerman and Judge, 2007) but can also be used for high-energy beta-emitting radionuclides such as ⁹⁰Y, although such measurements are more challenging (Pearce et al, 2007; Fenwick et al., 2014). In these cases, the IC response strongly depends on the transport of electrons in the radioactive solution and surroundings.

A few years ago, measurements of gaseous ²²²Rn samples contained in sealed glass ampoules, were performed with ICs at different NMIs and at the BIPM, in the context of a comparison using the SIR (BIPM.RI(II)-K1.Rn222). This comparison has highlighted relative differences of almost 5 % between laboratories and the results were not consistent within their uncertainties (Michotte et al., 2012). This issue has not yet been solved and gas glass ampoules are still being used to compare radon standards for submissions to the SIR. More recently, bias in measurements of gaseous ²²²Rn using ICs were investigated at the LNHB (Pierre et al., 2018). In this previous work, several measurements were carried out at the LNHB and at the BIPM and the variability of the response of ICs was investigated by considering the volume of the ampoule, the position of the sealing point and the thickness of the ampoule's base, which does not play a major role. The conclusion of this study highlighted that the variability of these gas glass ampoules could induce a measurement bias higher than the uncertainty of standard sources. Discrepancies observed were then interpreted to be the consequence of the contribution of high-energy electrons released from the main β emitting daughters of ²²²Rn, in particular ²¹⁴Bi (maximum beta energy of 3.27 MeV). These electrons have enough energy to reach directly the counting gas of the chamber (Michotte at al., 2006).

In order to address this phenomenon, we present here an additional study based on the Monte Carlo simulation of the Vinten type 671 IC (the IC used at the LNHB for standard transfers) with ²²²Rn gas glass ampoules. It has to be noted that the ionization of the IC gas is mainly due to the gamma emissions above a certain energy threshold (about 25 keV for the Vinten Chamber). However, high-energy electrons released by the ²²²Rn daughters such as ²⁴¹Bi or ²¹⁰Tl may contribute to the IC response via the Bremsstrahlung photons created in the solution itself, the container (the ampoule in this case), the holder walls and the inner wall of the chamber. In addition, electrons with the highest energies can reach directly the IC gas volume and interact with a high probability.

2. Methods

2.1 Decay scheme of radon

²²²Rn is a radioactive noble gas decaying through alpha transition (Fig. 1) to short half-life solid progenies and is one of the main sources of natural radioactivity.

National standards of ²²²Rn are available in several countries and comparison of these standards is necessary to ensure the international traceability of ²²²Rn measurement and to support the Calibration and Measurement Capabilities (CMCs) of the National Metrology Institutes.

The equilibrium with short-lived progenies (except for ²¹⁰Pb and daughters) is obtained after 4 hours. Table 1 shows the ratio between ²²²Rn and its daughters after 4 hours, having reached equilibrium.

2.2. Description of the simulations

The dependence of ²²²Rn results on ampoule geometry was studied by means of MC simulations of the LNHB Vinten chamber, based on original work by De Vismes and Amiot (2003) and Amiot (2004). The Vinten IC, designed on the basis of the IG42 IC constructed by Centronic (Woods et al., 1983), is used in routine at LNHB as a transfer instrument. The IC is composed of a cylindrical aluminium chamber (with a 10.5 L effective volume filled with N₂ at 1 MPa) with a coaxial re-entrant well, made of aluminium. The IC ionization current is collected via a central aluminium alloy electrode. The chamber is housed in a locally made shielding in order to reduce the background. The PENELOPE simulation model of this chamber (geometry and sample-to-detector configuration) has been well-validated

through different comparisons and for several gamma-emitting radionuclides (Amiot et al., 2012). The geometry used in our work is shown in Fig. 2.

Simulations were performed using PENELOPE code (version 2014) (Salvat, 2015; Baró, 1995) and Geant4 code (version geant4.10.0) (Agostinelli et al., 2003), studying the influence of the ampoule wall-thickness and the type (glass or metal), focusing on the ²¹⁴Bi daughter nuclide.

For a given radionuclide, the calibration coefficient (C_f in A/MBq) can be deduced from the total energy deposited in the gas E_d (eV) (obtained with PENELOPE calculations) according to the expression: $C_f = e \ge E_d / W$, where e is the electron charge and W the mean energy needed to produce an ion pair in the IC counting gas. Therefore, the variability observed in E_d reflects the variability of the calibration coefficient.

Usually, the calibration is carried out with glass gas ampoules with a wall thickness of approximately 1.3 mm, filled with ²²²Rn gas at very low pressure. It should be noted that for the SIR comparison BIPM.RI(II)-K1.Rn-222 (Michotte et al 2012), the ampoules submitted to the BIPM were all filled with a very low pressure. No specification was requested from the ampoule's supplier. As it was noticed that the thickness could be thicker than the nominal value, the simulations were performed at different thicknesses from 1.0 mm to 1.9 mm in steps of 0.1 mm.

2.3 ²²²Rn simulation

As it is very difficult and very time consuming to simulate the total disintegration chain of 222 Rn, and also because the IC instruments are not sensitive to alpha-emissions, the simulations were focussed on the daughters 214 Bi and 210 Tl radionuclides, which are assumed to influence IC response due to their high-energy β -emissions (Table 1).

The emitted particles by the selected daughter radionuclides are randomly generated as a uniform distribution in the whole volume of the radon source. In practice, when filling the radon ampoule, the solid daughters are on the ampoule walls, but considering the negligible pressure in the ampoules, using the volume source has no impact on the results. The initial electron energies emitted in the case of the beta transitions were taken from spectra calculated with the BetaShape program (Mougeot, 2016). Simulations were performed with a large number of primary events (about 10⁸ histories) in order to obtain good statistical precision of the result (i.e., the total energy deposited in the counting gas region deduced by averaging over all histories), below 1 %.

3. Results and discussion

Fig. 3 presents the average total energy deposition in the IC gas region (corrected by the ratios reported in Table 1) when we simulate only the β -transition emissions of ²²²Rn daughters, for glass ampoules with a nominal wall thickness of 1.3 mm. For Bi, Tl, Pb we used the total beta spectrum resulting from all the transitions. In this respect and considering the ratios given in Table 1, we can consider that ²¹⁴Bi represents the main daughter able to introduce a non-negligible contribution of direct ionization of the counting gas through its β -emissions.

Simulations were then focused on ²¹⁴Bi by considering individually the branches $\beta_{0,0}$ (3.27 MeV with a probability of ~19.6 %) and $\beta_{0,11}$ (1.5 MeV with a probability of ~17.5 %), and, finally, the total spectrum of all transitions.

The plot in Fig. 4 displays the variation of the total mean energy deposited in the IC gas region per decay as a function of ampoule thickness with the 214 Bi total beta spectrum. Only statistical uncertainties of the MC simulations are considered here in the plot (~ 0.3 %). A

decrease of the energy deposition in the chamber sensitive volume according to the ampoule thickness is clearly observed. This correlation yields a constant difference of about 14 % when we increase the thickness in steps of 0.1 mm.

To verify these results, the simulations were reproduced with another high-energy β -emitting daughter in the ²²²Rn decay chain, ²¹⁰Tl. The individual branches $\beta_{0,3}$ (~ 4.3 MeV with a probability of ~31 %) and $\beta_{0,2}$ (~ 4.4 MeV with a probability of ~13 %) as well as the total spectrum were considered. For the total spectrum, the observed behaviour (Fig. 4) is comparable to that of ²¹⁴Bi. The simulations for ²¹⁴Bi as well as ²¹⁰Tl (total β spectra) also show that the contribution of β transitions can be reduced to nearly zero when a 1 mm thick copper attenuator is positioned around the ampoules (Fig. 4 and 6).

For the individual branches $\beta_{0,3}$ and $\beta_{0,2}$ of ²¹⁰Tl, the same behaviour as for the total spectrum can be observed (Fig. 5). A similar behaviour was observed for the individual transitions of ²¹⁴Bi (not shown).

To investigate the influence of the nature of ampoules, we have also performed similar simulations using aluminium ampoules instead of glass, with the same variation of thickness. This resulted in a comparable behaviour of aluminium albeit at higher attenuation as one can see in Fig. 6 for the total β spectrum of ²¹⁴Bi. Again, a comparable behaviour of an aluminium versus a glass ampoule was observed for ²¹⁰Tl (not shown).

In view of these results, we can highlight that the contribution of the high-energy electrons released from ²¹⁴Bi decays significantly depends on the ampoule wall-thickness: a 0.1 mm variation in the wall thickness variation results in a 14% relative variation of the total energy deposited in the IC gas volume by the electrons. Even if this variation must be put in perspective with the response of IC to all the gamma emissions of the ²²²Rn daughter nuclides

not considered in the present study, this sensitivity may well be at the origin of discrepancies observed in the SIR.

4. Conclusions

In the context of the SIR, ICs are used to compare ²²²Rn standards. In the past, when using gas-filled glass ampoules, discrepancies between measurements remained unexplained (BIPM.RI(II)-K1.Rn222). In this work, the problem of the uncertainty of IC response due to the variability of the original glass ampoules used for ²²²Rn measurements was investigated by means of MC calculations. The objective was to check if the ampoule thickness could influence the response of the IC, by considering the high-energy electrons of the β transitions from the ²²²Rn decay chain radionuclides. It must be highlighted that Pearce et al. (2007) mentioned that "the simulation of IC response curves to beta emission is very sensitive to internal geometry of the IC and of the source, especially when the electron energy is high enough to cross all the layers and reach the gaseous sensitive volume". The results of the MC simulations of some ²²²Rn daughter radionuclides highlight a systematic and non-negligible correlation between the IC gas ionization and the variation of the wall thickness of the ampoules used in measurements. As consequence of this work and previous studies, a possible solution to reduce the high discrepancies, observed for instance in the frame of the SIR comparison, would be to use a specialist container for ²²²Rn that could be circulated between the participating laboratories. This ampoule can be made of Al with an optimised geometry (Bailat, 2016) in order to preserve the integrity of the sample during different displacements. As far as everyone would use the same ampoule, the properties of this ampoule do not need to be studied. If the glass gas ampoules are still to be used for comparisons, it could be useful to add to the protocol that a copper attenuator should be

placed around the ampoule. It must be noted that the pressure effect of ²²²Rn ampoules has not been investigated in this study but could also be a source of uncertainty. However, the submitted ampoules during the BIPM.RI(II)-K1.Rn222 comparison were approximately at the same low pressure, therefore the contribution of this should be significantly less than the 5% discrepancies observed.

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Figure captions:

Fig. 1: Decay scheme of ²²²Rn.

Fig. 2: View of the geometry of the chamber modelled. The hatching indicated by letter A corresponds to the radon source.

Fig. 3: Average of total energy deposition in IC gas region for β^{-} transition emissions of the ²²²Rn daughters, plotted on a log scale. This plot takes into account the ratio reported in Table 1.

Fig. 4: Variation of the mean energy deposited in the IC counting gas region per decay as a function of the radon ampoule wall thickness (with and without 1 mm copper attenuator), for the total spectrum. This plot does not take into account the ratio reported in Table 1.

Fig. 5: Average of total energy deposition in the IC counting gas region per decay for the transitions $\beta_{0,2}$ and $\beta_{0,3}$ of ²¹⁰Tl. This plot does not take into account the ratio reported in Table 1.

Fig. 6: Variation of the mean energy deposited in the IC counting gas region per decay as a function of the radon ampoule wall thickness (in glass or aluminium) for ²¹⁴Bi and ²¹⁰Tl (with and without 1 mm copper attenuator), for the total spectrum. Simulations made with Geant4. This plot does not take into account the ratio reported in Table 1.

Table captions:

Table 1: Activity ratio between ²²²Rn and progenies after 4 hours and major modes of decay.

Fig. 1.







Fig. 3.



Fig. 4.







 214_{Biβ} (max=3.3 MeV)/Glass-ampoule
with 1-mm-Cu attenuator $^{214}\textsc{Bi}_{\beta}\sc{-}$ (max=3.3 MeV) / Al-ampoule with 1-mm-Cu attenuator 8000 Δ Mean energy deposited in IC-gas region (eV) 210 -TIβ (max=4.4 MeV) / Glass-ampoule with 1-mm-Cu attenuator 210 TIβ⁻ (max=4.4 MeV) / Al-ampoule
with 1-mm-Cu attenuator 6000 4000 2000 1000 800 600 400 ø Ð θ 200 Ô Ø 0 e 1.2 1.3 1.4 1.5 1.6 2.0 0.9 1.0 1.1 1.7 1.8 1.9 Ampoule thickness (mm)

Table 1

Radionuclide	Daughter/ ²²² Rn	Mode of decay	Energy (keV)	Intensity (%)
²¹⁸ Po	1.00E00	$\beta_{0,0}^-$	260(12)	2.2 (3)
²¹⁴ Pb	1.00E+00	$\beta_{0,0}^-$	1019 (11)	9.2 (7)
		$\beta_{0,4}^-$	724 (11)	41.09 (39)
		$\beta_{0,3}^-$	667 (11)	46.52 (37)
		$\beta_{0,7}^-$	485 (11)	1.047 (17)
		$\beta_{0,9}^-$	180 (11)	2.762 (22)
		$\gamma_{5,0}$	351.932 (2)	35.60 (7)
		$\gamma_{4,0}$	295.224 (2)	18.414 (36)
		$\gamma_{4,1}$	241.997 (3)	7.268 (22)
²¹⁴ Bi	1.00E+00	$\beta_{0,28}^-$	822 (11)	2.76 (6)
		$eta_{0,21}^-$	1066 (11)	5.642 (43)
		$eta_{0,18}^-$	1151 (11)	4.339 (18)
		$eta_{0,16}^-$	1253 (11)	2.449 (10)
		$\beta_{0,12}^-$	1423 (11)	8.147 (28)
		$\beta_{0,11}^-$	1506 (11)	17.494 (36)
		$\beta_{0,6}^-$	1727 (11)	3.12 (4)
		$\beta^{0,4}$	1892 (11)	7.45 (5)
		$\beta^{-}_{0,0}$	3270 (11)	19.67 (20)
		$\gamma_{1,0}$	609.312 (7)	45.49 (19)
		$\gamma_{4,1}$	768.356 (10)	4.892 (16)
		γ _{6,1}	934.061 (12)	3.10 (1)
		$\gamma_{9,1}$	1120.287 (10)	14.91 (3)
		$\gamma_{12,1}$	1238.111 (12)	5.831 (14)
		$\gamma_{4,0}$	1377.669 (12)	3.968 (11)
		$\gamma_{11,0}$	1764.494 (14)	15.31 (5)
		$\gamma_{21,0}$	2260.3 (2)	4.913 (23)
²¹⁰ Pb	4.66E-04	$\beta_{0,1}^-$	17.0 (5)	80.2 (13)
		$\beta_{0,0}^-$	63.5 (5)	19.8 (13)
²¹⁰ Bi	1.14E-04	$\beta_{0,0}^-$	1162.1 (8)	99.99986 (2)
²¹⁰ Po	7.87E-7	$\alpha_{0,0}$	5304.33 (7)	99.99876 (4)

		$\beta_{0,11}^-$	1380 (12)	2
		$\beta_{0,10}^-$	1603 (12)	7
		$\beta_{0,9}^-$	1860 (12)	24
²¹⁰ Tl	2.12E-04	$\beta_{0,8}^-$	2024 (12)	10
		$\beta_{0,7}^-$	2413(12)	10
		$\beta_{0,3}^-$	4290 (12)	31
		$\beta_{0,2}^-$	4386 (12)	13
²⁰⁶ Hg	8.85E-12	$\beta_{0,0}^-$	1308 (20)	62 (7)
²⁰⁶ TI	1.68E-10	$\beta_{0,0}^-$	1532.4 (6)	99.885 (14)

<u>Highlights:</u>

- Simulation of the LNHB Vinten ionization chamber for ²¹⁴Bi and ²¹⁰Tl beta transitions.
- High-energy electrons from beta transitions can directly interact with the IC counting gas.
- IC response very sensitive to the ²²²Rn ampoules wall thickness.
- This phenomenon may explain 5% discrepancies observed in the ²²²Rn SIR comparison.