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SIMULATION OF THE RESPONSE OF AN IONIZATION CHAMBER TO ^{214}Bi AND ^{210}Tl EMISSIONS. APPLICATION TO THE MEASUREMENT OF ^{222}Rn.

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PENELOPE simulations of an LNHB ionization chamber (Vinten IC type 671) were performed to investigate the influence of glass ampoules used in ^{222}Rn standardization. Simulations were focused on the main β^--emitting daughters ^{214}Bi and ^{210}Tl (in secular equilibrium with ^{222}Rn). The emitted β energies being high, the IC response in terms of the measured current (directly proportional to the total energy deposited in the IC gas region) strongly depends on the transport of the electrons in the radioactive solution and surroundings (glass ampoule, chamber liner supports, …). The simulation reveals a non-negligible variation of the energy deposited in the IC gas region when considering the β transition emissions of the ^{222}Rn daughters. This reinforces the proposal for using a unique ampoule (made of metal to preserve the integrity of the sample) that will circulate between the metrology laboratories in the case of inter-comparisons.

Context

The degrees of equivalence of LNHB and IRA obtained with the SIR are not consistent with the results of the direct measurement comparisons ➔ *Fig. 1, Metrologia (2012), 49, Tech. Suppl. 06001* and *Tab. 1*.

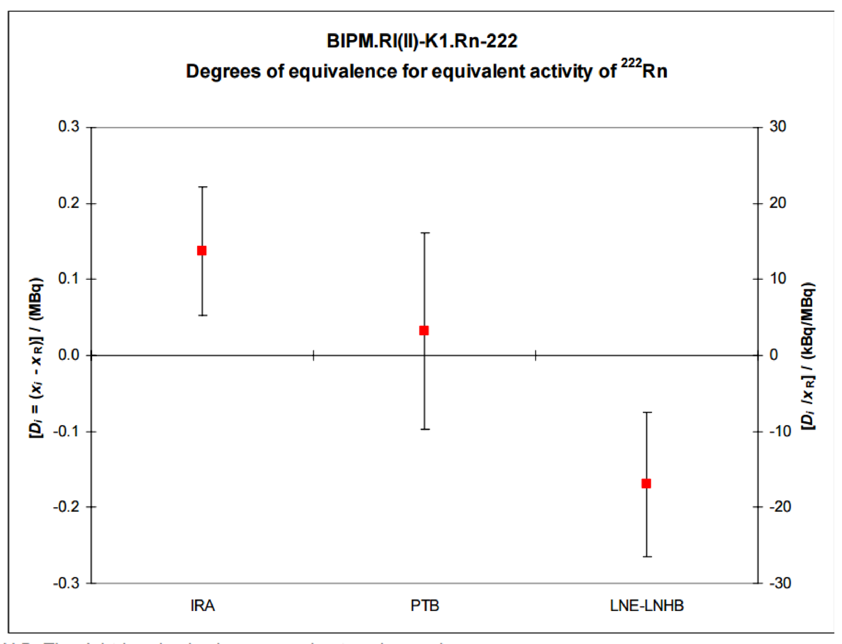


Fig. 1: Degrees of equivalence with the KCRV for ^{222}Rn

	Activity measured at LNE-LNHB (kBq)	Activity measured at IRA (kBq)
Standard made by IRA 11/9/12 12:00 UTC	256.6 (11)	256.8 (11)
Standard made by LNE-LNHB 11/9/12 8:00 UTC	224.7 (10)	224.2 (9)

Tab. 1: Comparison IRA and LNE-LNHB with metallic containers

➔ This problem was attributed to the effect of the BIPM glass ampoule wall thickness inhomogeneity
(Pierre, S., et al., 2018. Bias in the measurement of radon gas using ionization chambers: Application to SIR, Appl. Radiat. Isot. 134, 13-17).

Equipment used at LNE-LNHB for the radon calibration

- ✓ At LNE-LNHB, the radon sources are standardized with a defined solid angle system (DSA), based on the counting of the alpha particles of a cryogenic source.
  Sabot, B., et al., Applied Radiation Isotopes (2016) 118, 167-174.
- ✓ Transferable standards can be enclosed in glass-sealed ampoules (*Fig. 2*) or in metal container (*Fig. 3*), after cryogenic gas transfer.
- ✓ The same DSA method is used at IRA *(Spring, P., at al., 2006. Nucl. Instrum. Meth. A, 568, 752-759).*
- ✓ Metal containers are more convenient and easier to handle than ampoules.
- ✓ But glass ampoules are required for SIR measurements with ionization chambers (ICs).

Fig. 2: BIPM glass ampoule

Fig. 3: Metal container

- One IC used at LNE-LNHB is a Vinten type 671 equipped with a re-entrant ampoule device *(Fig. 4)*.

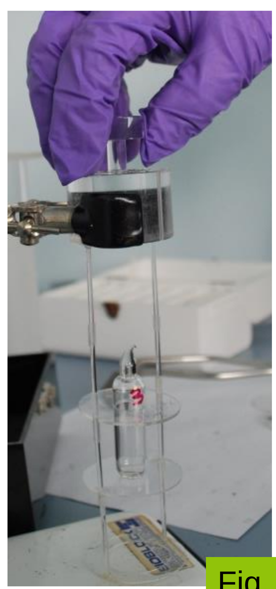


Fig. 4: Well-type re-entrant ampoule device in 2A IC

- This Vinten IC was simulated in this work (*Fig. 5*).
- The radon inside the ampoule is in dark blue in the center.
- The IC is composed of a cylindrical aluminium chamber (with a 10.5 L effective volume filled with N_2 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 locally made shielding in order to reduce the background.

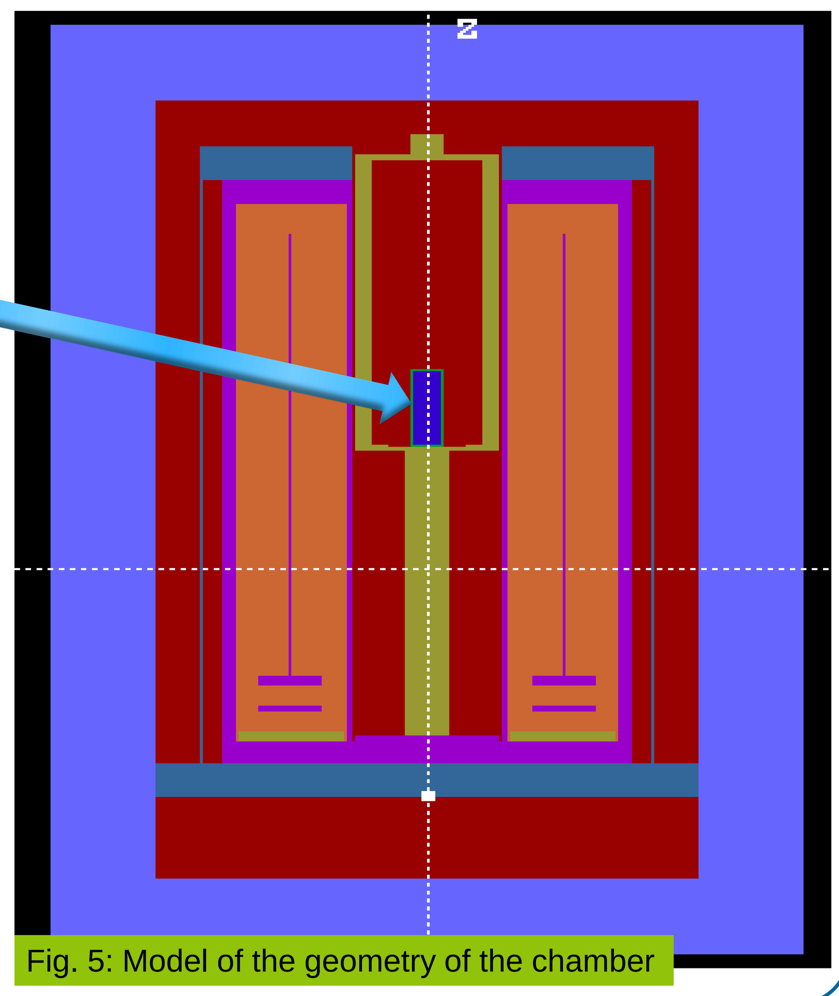


Fig. 5: Model of the geometry of the chamber

Simulations

- The calibration coefficient (C_f in A/Bq) can be deduced from the total energy deposited in IC gas E_d (obtained with MC simulation):

  $C_f = e \times E_d / W$ | e = electron charge | W= mean energy needed to produce ion pair in the IC gas

  ➔ variability observed on E_d reflects calibration coefficient
- As it is difficult to implement the total decay chain of ^{222}Rn *(Fig. 6)*, simulations were focused on the main β^--emitting daughters ^{214}Bi and ^{210}Tl *(Fig. 7 and 8)*.
- The mean thickness of the BIPM glass ampoule: 1.3 mm.
- As the wall can be thicker, simulations are performed from 1.3 mm to 1.9 mm, with steps of 0.1 mm.
- Emitted particles are randomly generated as a uniform distribution in the whole volume of the gas region.
- Initial beta electron energy distributions are calculated with the BetaShape program (*Mougeot, X., 2016. Appl. Radiat. Isot. 109, 177-182*).
- Large number of primary events generated (10^8) to ensure small statistical fluctuations of the results (i.e. the total energy deposited in the gas region deduced by averaging over the total histories), below 1%.

Particle transport in IC

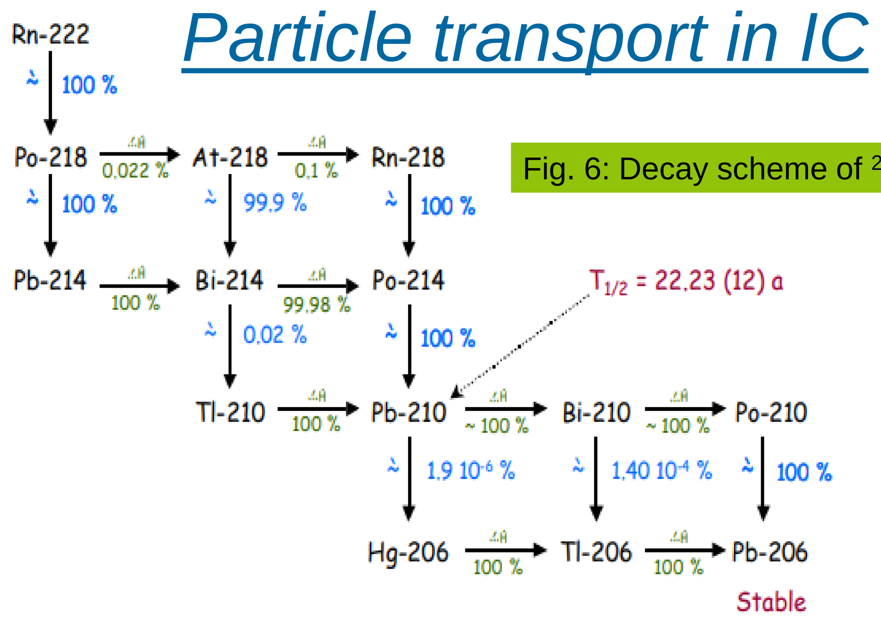
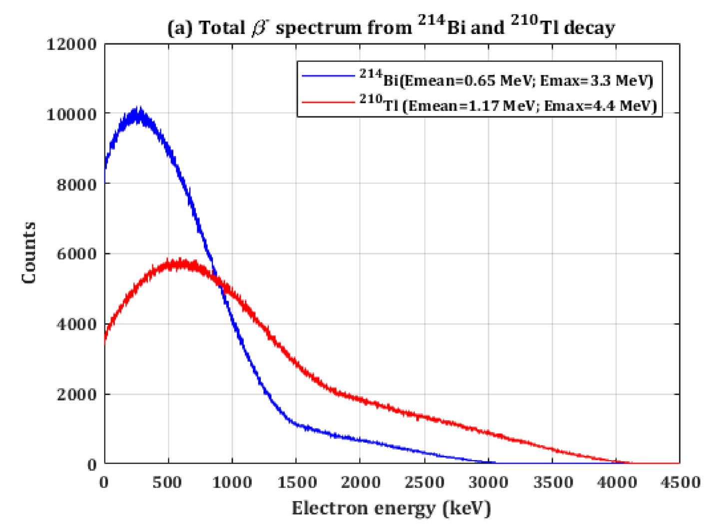


Fig. 6: Decay scheme of ^{222}Rn



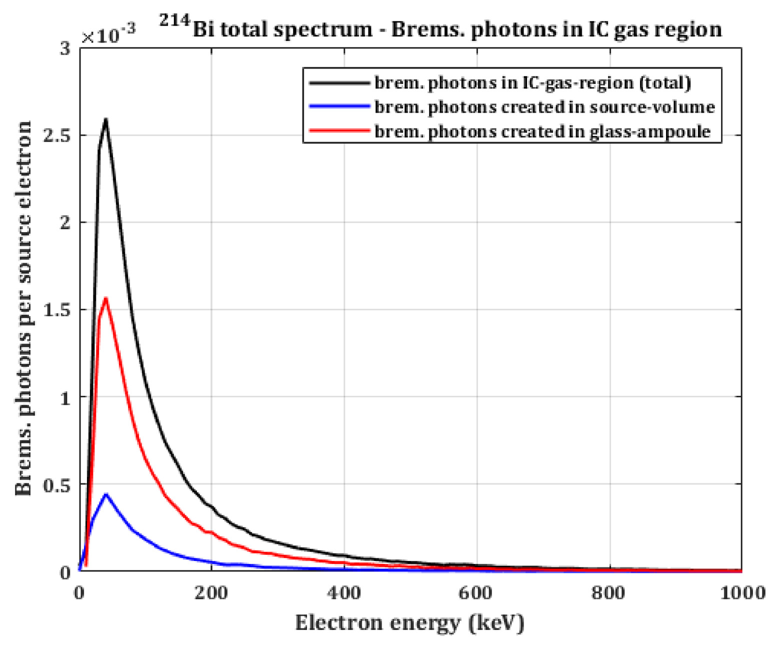


Fig. 7: Bremsstrahlung photons in IC –gas-region for ^{214}Bi

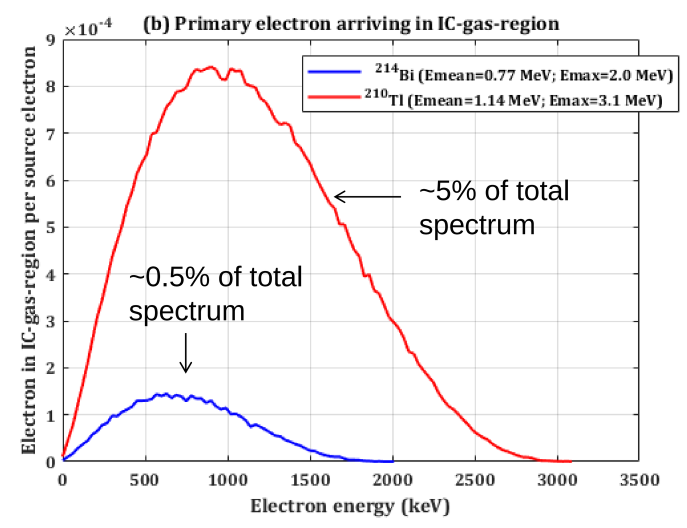


Fig. 8: (a) Primary electron released in the source from the total β spectrum of ^{214}Bi and ^{210}Tl
(b) Primary electron released in the source and arriving in IC gas region

Ampoule thickness variability

Fig. 9 shows the variation of the mean energy deposited in IC gas as a function of ampoule thickness.

➔ an increase of 0.1 mm of the ampoule thickness leads to:
✓ a decrease of 14% of the total mean energy deposited in the IC gas region for ^{214}Bi (for individual β transitions spectra and for average spectrum of β transitions).
✓ a decrease of 8% in the case of ^{210}Tl.
➔ same simulations were carried out with 1-mm-Cu attenuator around the ampoule: a significant reduction of energy deposited in the IC gas region was obtained.
➔ same behaviour was observed with metallic Al ampoule.
➔ consistent results are observed with both Monte Carlo codes used: PENELOPE and Geant4.

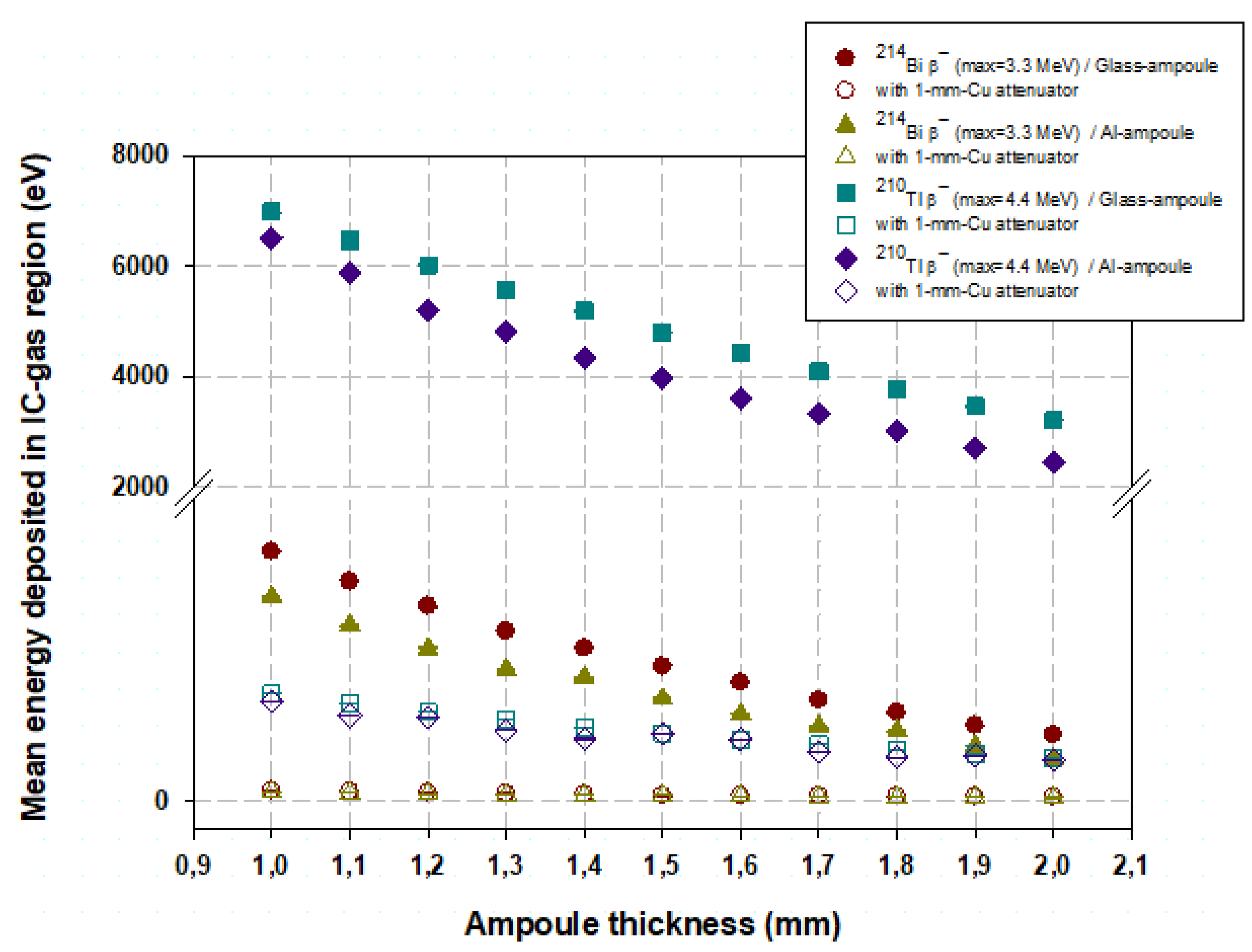


Fig. 9: Ampoule thickness variabilities

Conclusion

- Simulations clearly show a systematic and non negligible correlation between the IC gas ionization and the variation of the ampoule-wall thickness (via the direct contribution of high-energy electrons emitted in the source or through Bremsstrahlung photons created in IC layers).
- This phenomenon can explain the discrepancies observed in the BIPM.RI (II)-K1.Rn222 SIR comparison and was stressed by Pearce et al, 2007, *Metrologia 44 S67-S70* .
- A copper sleeve could be used to reduce discrepancies (at the price of the reduction of the sensitivity).
- The best solution would be to use a unique metallic container (made of Al for instance) for the SIR comparisons.