Protocol for a CCRI(II) Key Comparison of ^{222}Rn

1 - Introduction

Radon-222 is a radioactive noble gas decaying through alpha transition to short half-life solid progenies and is one of the main sources of natural radioactivity. It is monitored with commercial detectors and devices to evaluate radon activity concentration in rooms, water or soil. National standards of ^{222}Rn are available in several countries and comparison of these standards is necessary to ensure the international traceability of this radionuclide and to support the CMC's of the National Metrology Institutes.

2 - Previous comparisons

International comparisons were organized by NPL in 1992 and 1994 [1]. At this time, ^{222}Rn activity references were based on ^{226}Ra source emanation or on gamma-ray spectrometry.

Other CCRI(II) comparisons are available through the SIR [2,3].

Only comparisons using glass ampoules have been carried out until now. In the case of SIR measurements, the variability of the thickness of these ampoules could be an additional cause of uncertainty. Therefore, a comparison of standards using a more reliable metallic container, would be of interest and is proposed and described in this protocol.

A study concerning the glass ampoules, usually used for the SIR is joined to this Protocol[4]. This study explains why metallic containers are used instead of glass ampoules in this comparison.

3 - Specificity of ^{222}Rn comparison

Due to the short half-life of this radionuclide (3,8232(8) d), the logistics is important and all precautions must be taken in advance to be sure that measurement systems are operational at the reception date. The shipment will be organized as an exempted parcel transportation which imposes two constraints: the activity must be lower than 100 MBq and the dose rate at the contact of the parcel must be lower than 5 µSv/h.

4 - Comparison protocol

4.1 - Participants: IFIN-HH (Romania), INMRI-ENEA (Italy), IRA (Switzerland), KRISS (South Korea) and the pilot laboratory LNE-LNHB (France).

Each participant will measure the radon activity in his own setup.

4.2 - Source composition: ^{222}Rn will be delivered in a metallic container with approximately 650 kBq at time of shipment by LNE-LNHB.

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[1] Dean J.C.J. and Burke M. An intercomparison of ^{222}Rn measurement systems in European Laboratories. NIM A 339, 1994 pp 264-270

[2] Metrologia 2012 **49** Tech. Suppl. 06001

[3] Metrologia 2005 **42** Tech. Suppl. 06003

[4] Private communication - Internal Technical Note : LNHB 2015/20

4.3 - Container: a metallic (stainless steel) container of 105 cm^3 will be supplied by LNE-LNHB. The ^{222}Rn source is to be transferred by each participant into their own measurement device. The internal pressure of the container will be about 10^{-2} Pa of nitrogen, the typical partial pressure of ^{222}Rn being negligible.

The container is described in *Appendix 1*.

The source preparation is described in *Appendix 2*.

4.4 - Measurand: it will be the total activity of ^{222}Rn in the container at the reference date.

4.5 - Reference date: 01st July 2015, 12:00 UTC but could be modified according the shipping schedule.

4.6 - Recommended nuclear data: Decay Data Evaluation Project, http://www.nucleide.org/DDEP_WG/Nuclides/Rn-222_tables.pdf

4.7 - Distribution: the containers will be shipped to the participants in the last two weeks of June.

Transport of the ^{222}Rn containers will be arranged by the LNE-LNHB using their normal shipment arrangements for exempted parcels. A list of requested information will be sent to participants in order to enable an easy shipment.

Immediately after receipt, the participating institute shall acknowledge the reception, check for any damage to the samples and report this to the LNE-LNHB.

The containers shall be sent back by the participants to LNE-LNHB as soon as possible after measurements.

4.8 - Schedule of the comparison:
Reporting deadline: 15th September 2015
Draft A sent to participants: 30th October 2015
Draft A acceptance deadline: 30th November 2015
Draft B sent to participants: 30th January 2015
Draft B acceptance deadline: 30th February 2016

The LNE-LNHB will be responsible for maintaining up-to-date key comparison status reports and will transmit them to the CCRI(II) Executive Secretary.

The costs associated with the organization of the comparison, preparation, calibration and shipment of the ^{222}Rn comparison containers will be borne by the LNE-LNHB.

4.9 - Expression of the results : LNE-LNHB will send a copy of its own results to the CCRI Executive Secretary prior to the receipt of any results from participants. The LNE-LNHB will explicitly notify to the participants that they can submit their results to the LNE-LNHB. Please do not submit any results until you have received the notification. All results, method of standardization, associated uncertainties and other additional details which may be requested shall be sent to the LNE-LNHB using the reporting forms that will be provided.

Participants shall supply a list and an evaluation of the principal components of the uncertainty budget based on the Guide to the Expression of Uncertainty in

Measurement[5]. In addition to the principal components of the uncertainty, common to all of the participants, individual institutes must add any other components they consider appropriate.

5 - Preparation of the report on the comparison

5.1 - Treatment of the participants' results : To compare the results, LNE-LNHB will normalize the participant results to the volume of the containers.

5.2 - Reports edition :

LNE-LNHB is responsible for the preparation of the report of the comparison. The report passes through a number of stages before publication which are here referred as Draft A and Draft B.

After reception of the explicit request to send the reporting forms to LNE-LNHB (see 4.9), the results will be kept confidential by the pilot laboratory until the Draft A is circulated to all the participants.

A result from a participant is not considered complete without an associated uncertainty and is not included in the draft report unless it is accompanied by an uncertainty supported by a complete uncertainty budget. Uncertainties are drawn up following the guidance given in this Technical Protocol and in the reporting forms.

If, on examination of the complete set of results, LNE-LNHB finds results that appear to be anomalous, the corresponding institutes will be invited to check their result for numerical errors but without being informed of the magnitude or the sign of the apparent anomaly. If no numerical error is found, the complete set of results is used to prepare the Draft A.

The Draft A report is prepared as soon as possible after all the results have been received from the participants. It includes the results, uncertainties, standardization methods and experimental details transmitted by the participants, identified by name.

Draft A of the report is sent to all the participants for comments, with a reasonable deadline for replies. The date at which this draft is sent to the participants is taken to be the end date for the comparison and is subsequently referred to as such.

If any controversial or contradictory comments are received by the LNE-LNHB, they will be circulated to all participants and discussion continues until a consensus is reached.

Draft A is still considered as confidential to the participants. Copies are not given to non-participants, and graphs or other parts of the draft are not used in oral presentations at an external conference without the specific agreement of all the participants. The results may be the subject of an internal report if they are shown in relative terms and the names of participants hidden. At this stage, a participant may publish experimental techniques of special interest or new developments of a measurement method made in the frame of the comparison, as long as no information or comments are made about the comparison results.

Note that once all participants have been informed of the results, individual results and uncertainties may be changed or removed, or the complete comparison abandoned, only

[5] Evaluation of measurement data – Guide to the expression of uncertainty in measurement. JCGM 100:2008

with the agreement of all participants and on the basis of a clear failure of the travelling standard or some other phenomenon that renders the comparison or part of it invalid.

An institute that considers its result unrepresentative of its standards may request a subsequent bilateral comparison with the LNE-LNHB or one of the other participants. This should take place as soon as possible after the completion of the comparison in progress. The subsequent bilateral comparison is considered as a new and distinct comparison.

In the event that there is disagreement concerning the results or the interpretation of the results of a key comparison, and the disagreement cannot be resolved by the participants, by the key Comparison Working Group or by the Consultative Committee, the matter is referred to the CIPM for decision.

On receipt of final comments from the participants, the LNE-LNHB will incorporate them into a revised Draft A which, once circulated and agreed by all the participants, will become Draft B. The LNE-LNHB will then complete the Draft B to include the preliminary key comparison reference value and degrees of equivalence.

The Draft B is circulated through the participants and is no longer confidential and may be the subject of a publication, with the exception of the section containing the proposals for the degrees of equivalence.

The Draft B shall be sent to the CCRI Executive Secretary for preliminary editorial revision and circulation first through the KCWG(II) for technical review, and second through the CCRI(II) for approval. Once it is approved it will become the Final Report of the comparison and will be uploaded into the KCDB and eventually published in the *Metrologia Tech. Suppl.* series.

The BIPM will assist as needed at any of the stages described above and would possibly link the results in the Final Report to the BIPM.RI(II)-K1.Rn-222, if relevant.

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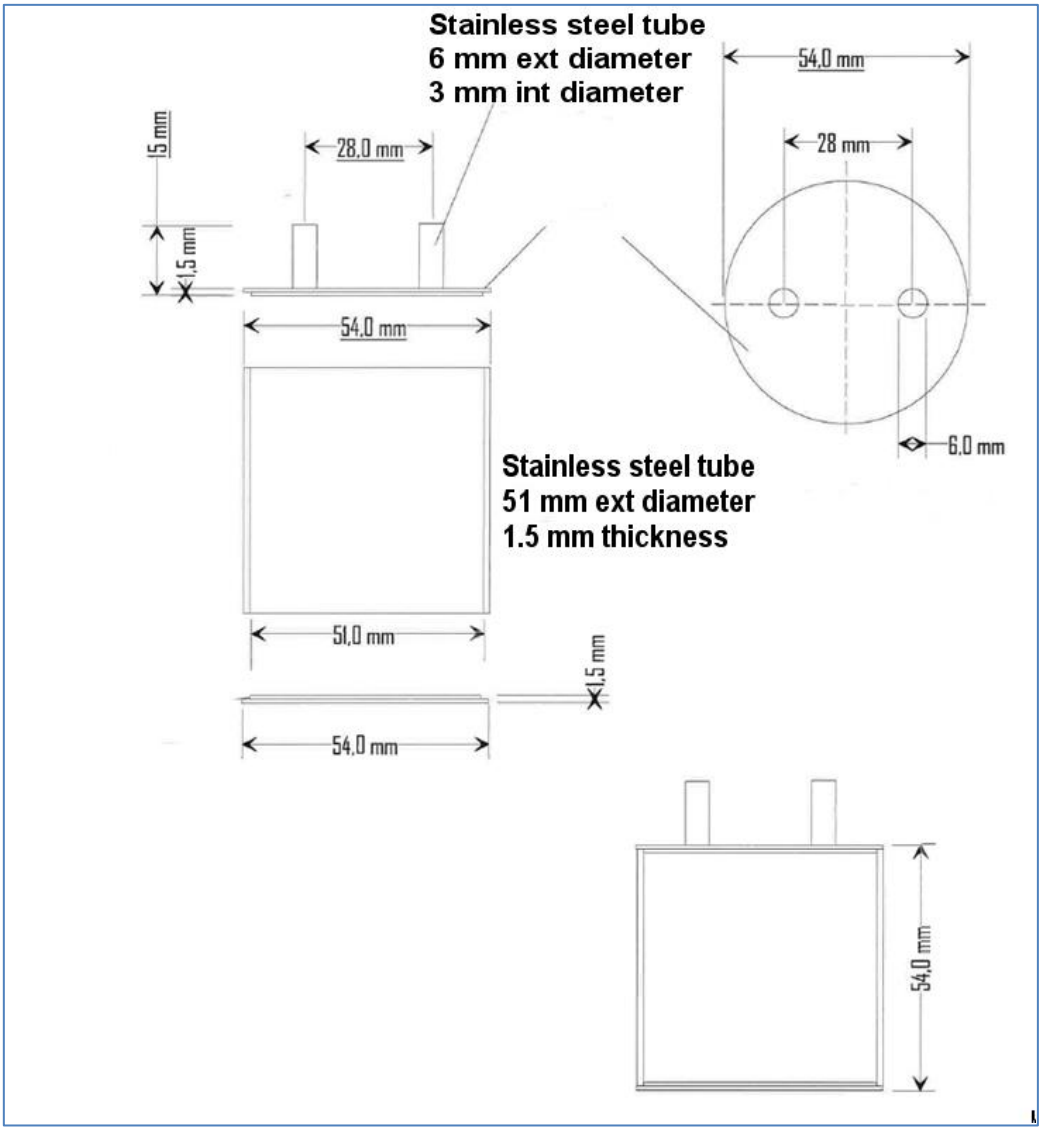
Appendix 1, container description



Appendix 2 – Containers preparation

The source preparation will be undertaken as follows: first a metallic container will be filled with 4 MBq of radon. Then, this container will be connected to 5 other containers to allow the homogeneous diffusion of radon between all the containers. Each participant will receive one of these 6 containers.

This principle is illustrated below for 3 containers.

Step 1: the metallic container 1 is filled with radon, the other containers and pipes are under vacuum.

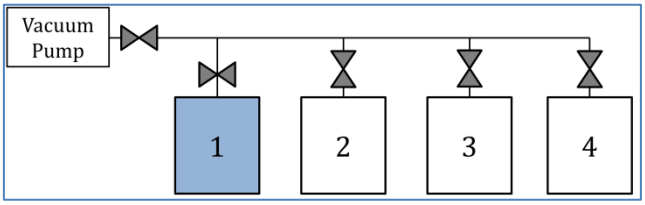


Figure 1: Source preparation method, step 1.

Step 2: the vacuum pump is isolated and all the containers valves are opened to allow the homogeneous diffusion of radon in the whole circuit.

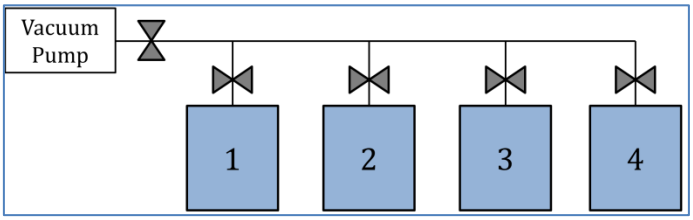


Figure 2: Source preparation method, step 2.

Result: the volume activity of radon is constant in the circuit and the quantity of radon in each container only depends on its volume, which was previously precisely measured at LNE-LNHB.

Each volume have been precisely measured at LNE-LNHB using a volume standard, a calibrated manometer and a calibrated thermometer, traceable to the French National

Metrology Institute for volume, pressure and thermometry units (Figure 3). The measurement method is based on the ideal gas law.

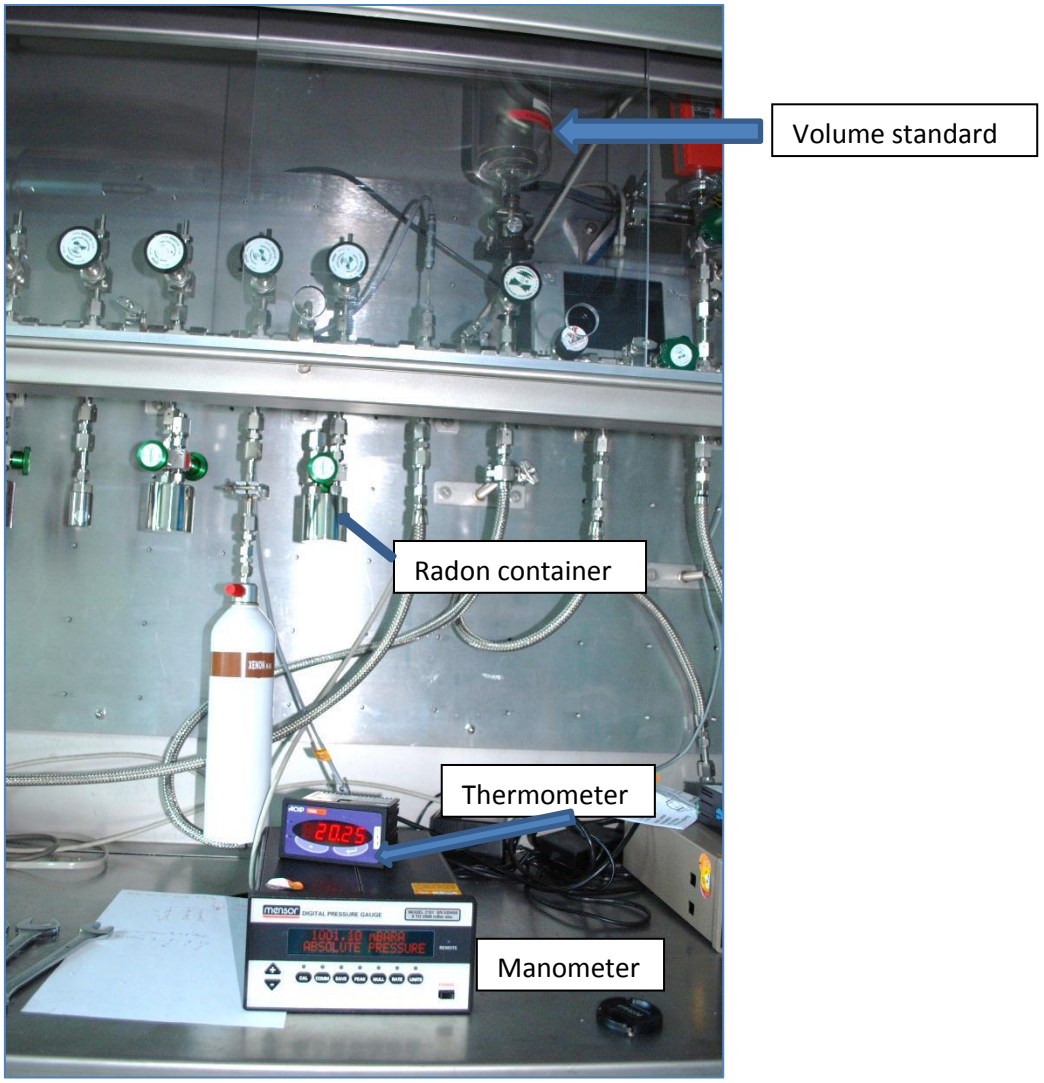


Figure 3: Device used to measure the volumes

Laboratoire National Henri Becquerel

LNE-LNHB

NOTE TECHNIQUE

Référence de la note	LNHB 2015/20

TITRE : Factors of influence in the measurement of radon activity standards using ionization chambers and gamma-ray spectrometers: a preliminary study

RESUME : Two main non-destructive techniques can be used to measure standard ^{222}Rn ampoules: ionization chambers and gamma-ray spectrometry, the former being used in the Système International de Référence (SIR) for international comparison purposes. These two methods are based on the detection of photonic emission (both from gamma emission and electron bremsstrahlung) and their reliability requires that the variability of the ampoules have a negligible influence on the detector response. This is why the influence of glass thickness and sealing point position on the detector response has been studied. The glass thickness is quantified by a photon absorption method using a ^{241}Am point source, and the global study of the detector response is quantified by measuring various standard ^{222}Rn ampoules with different glass thicknesses, internal volumes and positions of the sealing point. This leads to the conclusion that the variability of the ampoules induces measurement differences higher than the uncertainty of the standard sources, showing that great care must be taken if these measurement methods are to be used for absolute activity determination.

Mots-clés : **^{222}Rn, ionization chamber, gamma-ray spectrometry, glass ampoules.**

0	13/4/2015	**Emission initiale**
IND.	**DATE**	**OBJET**

	Auteur(s)	Vérificateur(s)	Chef de laboratoire	Chef du LNHB
Noms	S. Pierre, B. Sabot, P. Cassette, S. Courte, L. Ferreux, I. Le Garrérès, V. Lourenço, G. Ratel, F. Rigoulay	B. Chauvenet	Thierry Branger	Loïc de Carlan
Dates	03/04/15		03/04/15	13/4/15
Signatures				

1. - INTRODUCTION

The system ensuring primary activity measurements of ^{222}Rn, based on the defined solid angle alpha counting (DSA) of a cryogenic solid source, developed in our laboratory [1] has recently been improved [2]. This system allows the production of a radon standard in a precisely manufactured metal container or in a flame-sealed glass ampoule. Metal containers are generally preferred as they are easy to handle and allow a non-destructive control of the source activity. This control can be carried out, after cryogenic gas transfer, by the DSA measurement, but also by gamma-ray spectrometry, using a calibrated spectrometer. Nevertheless, preparation of radon standards in flame-sealed ampoules is sometimes necessary, especially for international comparisons within the Système International de Référence (SIR) of the Bureau International des Poids et Mesures (BIPM). In this system, which is the cornerstone of the international traceability of standardizations of γ-ray emitting radionuclides, and therefore of radon standards, the ampoule is placed inside a well-type re-entrant ionization chamber (IC), producing a current which is compared to that obtained for a very stable radium source. The computation of the ratio between these two currents enables the evaluation of a so-called equivalent activity characterizing the radionuclide under study. The use of glass ampoules raises two important questions:

"What is the influence of the position of the sealing point on the response of the ionization chamber?", which is known to be very sensitive to the geometry of the source and

"What is the influence of the variation of the thickness of the glass ampoule on the response of the ionization chamber?".

To address the first point, and evaluate the possible variation of the ionization chamber response, radon standards in glass ampoules were prepared by voluntarily changing the position of the sealing point and then measured in three ionization chambers with different wall materials and thicknesses. The second point was studied by using glass ampoules with various glass thicknesses. These ampoules were selected from a batch, by measuring the thickness of the wall using a radiation absorption method developed in our laboratory and the response of the ionization chambers were correlated with the thickness of the ampoule.

This whole study allows the determination of an uncertainty component due to the variability of the ampoules used for SIR measurements of radioactive gases and more generally for any measurement method based on the detection of the gamma emission of radon and progeny.

2. - METHODOLOGY

Three SIR ampoules were selected, out of a batch of nineteen, to maximize the variability of their thickness and volume. The volume of each ampoule was precisely measured using distillated water and a weighing method and equations were derived to deduce the effective volume of the ampoule from the position of the sealing point. The three ampoules were filled with the same activity of ^{222}Rn. One of them was sealed at a different position, and another one had a thick glass bottom... All the ampoules were measured in ionization chambers, and then standardized by DSA after destruction.
Three different ionization chambers were used: two chambers (named 2A and 6D) at LNE-LNHB and the BIPM SIR chamber number 389 [3]. These chambers are slightly different. The chambers 2A and 6D are made of an alloy of aluminium, and have volumes of 8 L and 1.3 L respectively. The thickness of their walls is 2 mm for 2A and from 0.8 mm to 1 mm for 6D. The BIPM chamber is made of steel. The reentrant tube made of mild steel, 0.5 mm thick, has an inner diameter of 25 mm. The sensitive volume of the BIPM SIR chamber is 5.2 L. Both 2A and BIPM SIR chambers contain nitrogen, whereas chamber 6D contains a mixture of 90 % argon and 10 % xenon. Chamber 6D is more sensitive to low energies. The ionization chamber current was normalized by the activity of each ampoule determined with the DSA method [1]. All these measurements were decay-corrected and expressed at a common reference date; the decay during the measurement has also been taken into account. Finally, a germanium detector was also used to confirm the general trends of the results.

3. - RESULTS

3.1 Influence of the thickness of the bottom

The responses of the three ionization chambers are presented in Figure 1. The characteristics of the three ampoules are presented in Table 1.

Table 1. Ampoule properties.

Number of ampoule	Volume and uncertainty/ mL	Properties
1	5.6413 (2)	Sealed with a long tube
2	5.5800 (2)	Sealed with a short tube
3	5.5493 (2)	Thick glass bottom, sealed with a short tube

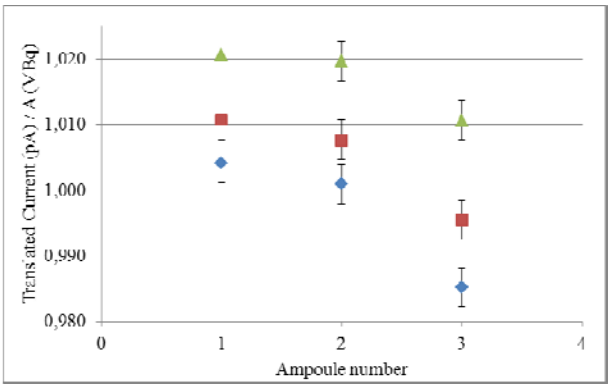


Figure 1. Response of each ionization chamber for each ampoule.

▲ 2A LNE-LNHB IC ■ BIPM IC ♦ 6D LNE-LNHB IC

It can be observed that the responses of the three chambers are similar. The ratios of these responses between pairs of ampoules are presented in Table 2 from which several conclusions can be drawn:

(1) There is no evidence of a large influence of the sealing position on the response, as the observed differences on the ratio 1/2 are within the uncertainties on the activities (about 0.3 %).

The ratio of the responses of ampoules 2 and 3, showing the influence of the glass thickness of the bottom of the ampoule, reaches 1 % or even 1.6 % for chamber 6D. This parameter thus plays an important role in the response of the ionization chambers.

Both effects are added, as observed in the ratio between ampoule 1 and ampoule 3. The relative deviation reaches 1.5 % for the SIR chamber and 1.9 % for LNHB's chamber 6D.

Table 2. Ratio of IC currents and uncertainties for pairs of sources.

Ratio	2A IC	6D IC	SIR IC
1/2	1.001(4)	1.003 (3)	1.003 (4)
1/3	1.010 (4)	1.019 (4)	1.015 (4)
2/3	1.009 (4)	1.016 (4)	1.012 (4)

3.2 Influence of the volume

The responses of the three chambers as a function of the internal ampoule volume are presented in Figure 2, where it can be observed that the influence of the volume on the response is similar for all three.

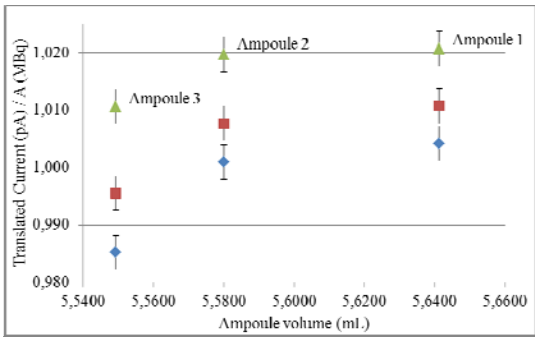


Figure 2. Chamber response for each ampoule volume.

▲ 2A LNE-LNHB IC ■ BIPM IC ♦ 6D LNE-LNHB IC

The volume difference between ampoules 1 and 2 is much larger than the volume difference between ampoules 2 and 3. Nevertheless the responses of ampoules 1 and 2 are very close to each other, while the response of ampoule 3, which has a thicker glass bottom, is significantly lower. This shows that the main parameter of influence seems to be the thickness of glass and not the internal volume of the ampoule. In order to more conclusive, a study with a larger batch of ampoules should be undertaken.

4. - GAMMA-RAY SPECTROMETRY MEASUREMENTS

4.1 Ampoule wall thickness measurements

A non-destructive photon-absorption measurement system was designed at LNE-LNHB in order to quantify the glass thickness of the ampoules. A thin metal rod with an electroplated ^{241}Am source is introduced inside the ampoule and the photo emission is measured using a High Purity Germanium detector (HPGe). The measurement of the absorption of three characteristic photon emissions of ^{241}Am, (21.16 keV (E1), 26.34 keV (E2) and 59.54 keV (E3)) is used to quantify the glass thickness with the Beer-Lambert exponential law. The position of the rod is reproducible in order to allow comparative measurements without ampoule and after changing the ampoules. The measurement device is shown in Figure 3.

Figure 3. HPGe detector with the ampoule and the ^{241}Am source.

The relative standard deviation of the counting rate is 0.3 % for E1, 0.4 % for E2 and 0.1 % for E3. The reproducibility is calculated from three measurements where the ampoule is removed between each measurement, leading to a relative standard deviation of 1 % for E3 and 3 % for E1 and E2.

The glass thickness of the bottom of six selected ampoules was measured. The results show that for the three energies of ^{241}Am, the absorption curves have the same tendency, which is presented, as an example, for energy E1 in Figure 4. This allows a classification of the ampoules *vs.* their bottom thickness. It was observed that for the six selected ampoules, the relative standard deviation on this parameter is approximately 26 %, which is significantly larger than the fluctuations due to the reproducibility of the measurement procedure. This means that there is a significant variability in the thickness of the glass of the ampoules used for the SIR.

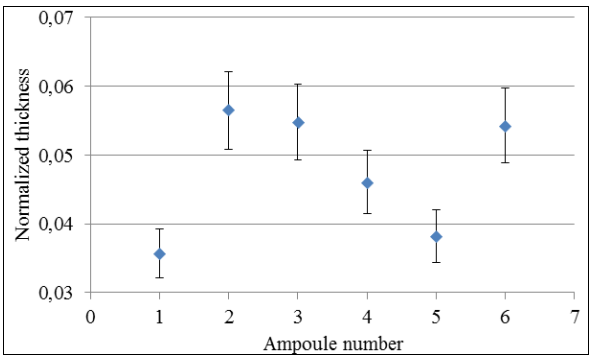


Figure 4. Response of HPGe at energy E1 for six ampoules.

4.2 Radon source measurements

After displaying the influence of the bottom thickness of the six ampoules with the ^{241}Am source, they were filled with radon and measured with a HPGe spectrometer. The results were then normalized with the radon activity obtained with DSA measurements. The measurement setup is shown in Figure 5.

Figure 5. Radon ampoule measurement setup with a HPGe detector.

The measurement concerns the gamma-ray peaks of ^{214}Bi (especially that at 609 keV), with all values decay-corrected. The dominant uncertainty contributions for the determination of the area of the 609 keV peak are the reproducibility (< 1 %) and the counting fluctuations (0.3 %). The decay-corrected counting rates are normalized by the sample activity of each ampoule. Coherent responses are observed using various ^{214}Bi energies and the results obtained for the 609 keV peak are displayed in Figure 6.

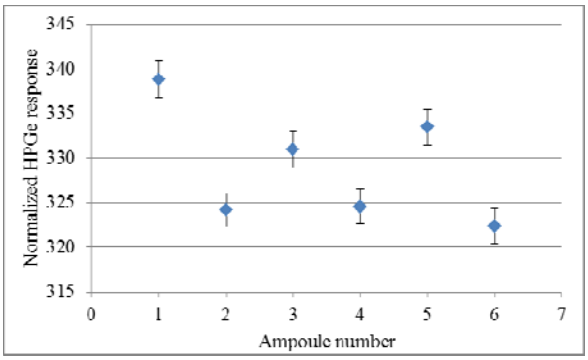


Figure 6. Decay corrected normalized counting rate of the ^{214}Bi 609 keV peak for each ampoule (k=1).

It can be observed in Figure 6, that some results are not consistent within the uncertainties. This confirms the previous observation that the response of gamma detectors depends on the thickness of the ampoule. Thus great care must be taken during ampoule selection when using gamma spectrometers to measure the activity contained in radon filled ampoules.

5 – CONCLUSION

The measurement of radon ampoules with ionization chambers is used to compare the radon standard of different laboratories of the BIPM in the framework of the SIR. However, it has been observed that the measurements made under the same conditions are not always compatible within uncertainties using three different ionization chambers in two different laboratories. The coherence of the measurements reported in this paper shows the possible influence of the thickness of the ampoule glass on the response of the chambers.

This implies that one must be very careful when assessing the uncertainties for such measurements, especially when this information is used for international comparisons of activity. Until now, no uncertainty was attributed to ampoule variability. This paper shows that a more systematic study on a large ampoule batch is necessary to derive a reliable uncertainty component related to ampoule variability.

A much better approach would be to define a reliable procedure to select the ampoules to be used for the SIR, in order to minimize the effect of the glass thickness. This procedure could be based on the measurement using ^{241}Am source as presented in this work.

REFERENCES

1 - Picolo, J.L., 1996. Absolute measurement of radon-222 activity. Nucl. Instrum. Methods A 369, 452-457.
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3 - Ratel G., The Système International de Référence and its application in key comparisons, Metrologia, 2007, 44(4), S7-S16.