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Design of a distributed radioactivity standard – feasible or not?

Final report on the project "realisation of the becquerel"

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Abstract

The "realisation of the becquerel" project is addressing the issue of potential failure of the SIR (Système International de Référence), by developing a fully reproducible ionisation chamber, which could serve as the realisation of the SI unit for radioactivity, the becquerel. To achieve this, all dimensional and operational parameters that influence the response, together with their tolerances should be investigated and brought together in a detailed construction plan. From 1997 until 2013, scientists and engineers from various National Metrology Institutes and the IRMM have studied the parameters of influence and have investigated many technical and operational issues, which are summarised in this report. Not compromising the requirement of a reproducibility of 0.2 % at 30 keV photon energy, only the highly expensive and dual-use material beryllium can be used for the construction of one of the most critical components, which makes it unlikely that such an ionisation chamber will ever be built.

1 Introduction

1.1 The international system of units

The international system of units (SI - Système international d'unités) comprises a coherent system of units of measurements built on seven base units and an unlimited number of derived units formed by powers, products or quotients of the base units. The definition of each base unit of the SI is drawn up so that it is unique and provides a sound theoretical basis on which the most accurate and reproducible measurements can be made[1].

The definition of SI base units evolves. For example, the original metre and kilogram, called the Mètre des Archives and Kilogramme des Archives, were constructed in 1799 to be one ten-millionth of a quadrant of the Earth and the mass of a cubic decimetre of water respectively. Physical artefacts to represent the reference for these units were created. These artefacts are prone to deteriorate over the years of use. For that reason, definitions that rely directly on stable physical properties are preferred. Today, the metre is defined as the length of the path travelled by light in vacuum during a time interval of 1/299 792 458 of a second. However, the unit of mass (the kilogram) is still defined as the mass of the international prototype of the kilogram. But, it is planned that in 2017 there will be a decision at BIPM to replace the kg with a value of the Planck constant measured using a so-called Watt-balance. This new definition is planned to enter into force in 2018.

National Metrology Institutes (NMIs) demonstrate the international equivalence of their measurement standards and the calibration and measurement certificates they issue. This system relies on the traceability to the SI units. This is also the case for the unit becquerel.

1.2 The becquerel

The becquerel (symbol Bq) is the unit of activity referred to a radionuclide. One becquerel is defined as the activity of a quantity of radioactive material in which one nucleus decays per second. The becquerel is therefore equivalent to one reciprocal second[2].

Due to the radioactive decay process, it is impossible to keep a standard for activity for all radionuclides (about 3000, of which around 100 are relevant). A pragmatic solution to this problem is the SIR (Système International de Référence).

1.3 The International Reference System (SIR)

The international reference system for γ-ray emitting radionuclides is relying on ionisations chambers which were calibrated over a long period of time with significant efforts. The best calibrated ionisation chamber to date is the SIR, operated by the BIPM (Bureau International des Poids et Mesures). The BIPM acts in matters of world metrology, particularly concerning the demand for measurement standards of ever increasing accuracy, range and diversity, and the need to demonstrate equivalence between national measurement standards [3].

Participating national and international metrology institutes submit their standardised radionuclides in glass ampoules to the BIPM where the ionisation current generated by the SIR is compared with the current obtained with a ^{226}Ra reference source. (^{226}Ra has a long half-life of 1600 (7) years[4].) About 900 of radioactive solutions have been measured for a total of about 60 radionuclides during the past 35 years. The ratio between the standardised massic activity and the SIR ionisation current is the basis on which the becquerel is realised and international equivalence is demonstrated.

1.4 Potential issues with the SIR

Should any of the critical components of the SIR ionisation chamber fail (either the chambers themselves or the reference sources) a significant amount of effort would be in peril and to some extent, the whole traceability chain in the ionising radiation metrology.

1.5 Objectives of the "realisation of the becquerel" project

The "realisation of the becquerel" project is addressing the issue of potential failure of the SIR, by developing a fully reproducible ionisation chamber. This reference instrument has an output (response function) which is equal within an allowed margin to any copy of such chamber. To achieve this, all dimensional and operational parameters that influence the response, together with their tolerances should be investigated and brought together in a detailed construction plan.

Once such a single reference instrument has been calibrated for a large number of radionuclides and the response curve is derived, all chambers built and operated according to the construction plan in an SI-traceable way (gas pressure, current measurement, dimensions, …) will have the same response within the agreed margin.

To foresee possible replacements it must be feasible to build another reference chamber, at any point in time and by any metrology institute. The use of a reproducible reference chamber by several institutes has also the major advantage to exchange directly applicable calibration information on new radionuclides, source containers, solutions, etc.[7] In particular for short-lived radionuclides, the availability of a common reference chamber in all parts of the world would enormously facilitate demonstration of international equivalence. Currently, activity standardised solutions of short-lived radionuclides cannot be compared with the SIR because of the distance and time needed for export and import of radioactive substances. The current procedures foresees in the use of a travelling transfer instrument (SIRTI).

1.6 History

This project was initiated by Dietmar Reher (IRMM), Mike Woods (NPL) and Bruno Denecke (IRMM) around 1997. A lot of scientific and technical research has been invested in the project and several National Metrology Institutes contributed to the project: NPL (UK), LNE-LNHB (France), PTB (Germany), NIST (US), NMIJ (Japan), CIEMAT (Spain). Investigations into materials, designs and construction methods began in 1997 and Monte-Carlo simulations were carried out[5] to assess the feasibility and the ease of construction.

The project was endorsed by the BIPM working group BqWG(II) – the becquerel at the basic level – where the findings were discussed at their regular meetings. One should point out that he BIPM has no resources to carry out such work itself. It is highly depending on the work carried out by NMIs in member state laboratories.

Over the years, it became clear that the objectives of the project are very difficult to achieve, if not technically impossible. Due to lack of financial and human resources, the NMIs one by one stopped supporting the project. Finally, also IRMM stopped performing research on the project in 2013.

1.7 Aim of this report

It would be a tremendous endeavour to compile a conclusive report on the project. Instead, this report aims to briefly summarise the research that has been performed, mainly after the publication of the report by Camps and Paepen[7], and to draw conclusions on the different design issues, taking into account the current state-of-the-art technology. For completeness, reports and publications not referred to in this report are listed after the references.

2 Requirements of the reference instrument

The broad goal of the project is to replace the dependence on a unique system based in one location, like the SIR, with a system that can be rebuilt identically by any skilled laboratory, anywhere and at any time in the future. The best approach was found to be the realisation of a new measurement system, based on an ionisation chamber, with characteristics similar to the SIR, but fully reproducible.

The deliverables of the project are a complete set of specifications concerning all the parameters of the system that have an impact on the measurement result. This includes the materials and their composition (including the gas), the dimensions of all the parts of the chamber (including tolerances), and the properties of the current measurement system. Procedures on how to build and assemble the parts, and how to operate the system shall be included.

The objectives result in a set of principal requirements (Table 1), which form the basis of the research topics discussed further.

Principal requirements for the design of a distributed radioactivity standard	
Reproducibility	The response shall be reproducible within 0.2 % for photons in the energy range from 30 keV to 2.6 MeV.
Traceability	It shall be possible to specify all values and tolerances of the parameters influencing the response in SI units and to demonstrate traceability.
Stability	The response shall be stable response over tens of years.
Response linearity	There shall be a linear behaviour of the current output versus activity.
Cost	Production and operational cost should be as low as reasonably possible.
Simple design	The design should be as simple as possible.
Ease of operation	Easy access for source positioning.
Beta emitters	Preferably, the chamber should allow measurement of pure beta emitters.

Table 1: Principal requirements

3 Key components

The key components of an ionisation chamber are depicted in Figure 1. An ampoule with a radioactive solution is placed in a well or inner tube. (The ampoule holder is not shown in the figure.) A pressure vessel consisting of the inner and outer tube, and top and bottom flanges holds a pressurised gas. The main part of the radiation passes through the ampoule wall and the well, and will ionise the gas: electrons and ions are created.

For a specific chamber filled with a certain gas pressure, the amount of ionisations is a function of the type and energy of the radiation (which depend on the nuclide) and the activity of the solution, which is the quantity of interest. In order to measure the number of ions and electrons, an electric field is applied by means of biased electrodes. The ions and electrons will drift in opposite direction under influence of the electric field, which is observed as an electric current, flowing between the electrodes. This ionisation current is measured outside the chamber.

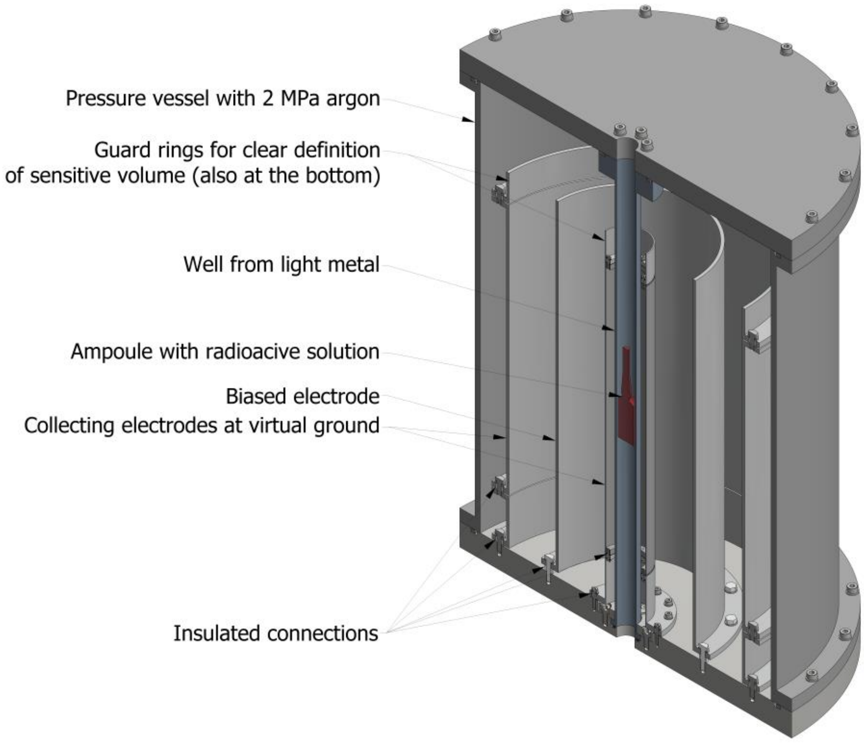

Figure 1: Cross section of the 2010 ionisation chamber design

4 Prototypes

A first prototype (Figure 2) was built in 2005[6]. It is a cylindrical ionisation chamber with a single electrode that collects the moving charges. A bias voltage is applied on the outside of the chamber. Extensive tests were performed with inner tubes made from Vespel® (a polyimide) coated with metal, and aluminium. Many of the operational and design parameters were investigated, such as the dependence of the response on the gas pressure the applied high voltage and the reproducibility of source positioning[7]. Identified issues with the first prototype are:

- The asymmetry of the electrode configuration;
- An improper design of o-ring grooves;
- The insufficiently reliable electric feedthrough, made in-house from sapphire and brass parts: the feedthrough started leaking gas, and the inner conductor broke off;
- The outside is at high voltage, requiring an extra cover for safety reasons, and for shielding the chamber from external electromagnetic disturbances;
- An asymmetric behaviour of the response as a function of the radial source position, most likely caused by a slight buckling of the Vespel® inner tube under the longitudinal compression stress applied by the flanges;
- A step in the response as a function of longitudinal source position, caused by a visually observable edge in the aluminium inner tube, due to improper machining;
- Electrons scattering back from outer tube have a large contribution to the response, causing an excessive dependency of the response on the deformation of the chamber under pressure and on tolerances and surface roughness[7].

These findings were used in the design of a second prototype (Figure 3).

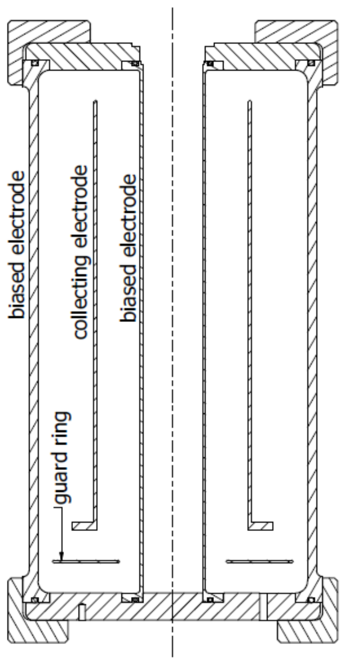

Figure 2: The first prototype, built in 2005.

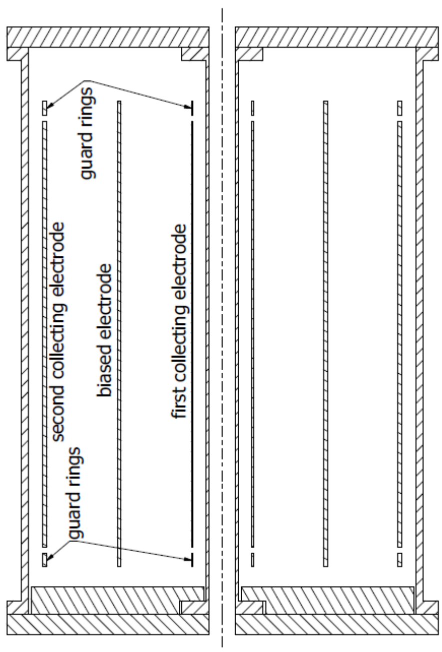

Figure 3: The 2010 prototype (never built).

The design of the second prototype was finalised in 2010. The cylindrical chamber contains three electrodes and four guard rings. The first and the third electrode are collecting electrodes (virtually connected to ground potential by the current measuring system), while the second, central electrode is the biased electrode. Each collecting electrode is shielded by two guard rings, one on the upper side and one on the lower side, separated from the collecting electrode by thin ceramic insulators. Alternatively, the reverse configuration where the second electrode serves as the collecting electrode should be considered as well, since electrons scattered back from the outer tube will be stopped in the third electrode. This is an unwanted effect that also applies to the inner tube and first electrode. (Back-) scattered electrons should preferably not hit the collecting electrodes and contribute to the collected current.

The major expected advantage of this design with respect to the first prototype is the precisely defined collection volume: a separate volume inside the chamber defined by the electrodes which do not have the structural task of holding the gas pressure[8]. In addition, this design is safer to operate since the outside of the chamber is electrically grounded. The 2010 design was never built.

5 Reproducibility

5.1 Requirement

The objective concerning reproducibility is that the response, of any ionisation chamber built according to specifications, for any photon in the energy range between 30 keV and 2.6 MeV, shall not differ by more than 0.2 % from the response of any other ionisation chamber built to the same specifications.

The response of the chamber is the electric charge generated by the chamber associated to one emitted photon of a given energy. A convenient unit to express the response is attocoulomb per photon (1 aC = 10^{-18} C), which translates to pA/MBq of activity for nuclides with 100 % emission probability for the photon with that energy.

5.2 Discussion

The requirement at the lower energy limit of 30 keV originates from the wish to be able to measure ^{125}I or similar medical nuclides with low photon energies. It is recognised that the 0.2 % reproducibility requirement at this energy is at the very edge of what is technically achievable. Even on a single chamber (the SIR), the spread amongst ^{125}I measurements is quite large [Michotte, BqWG(II) 13/05/2013].

The reproducibility at the lower energy limit is the most demanding requirement. At the BqWG(II) meetings, several times it was discussed to (at least partly) give up the extension to lower energy. However, the working group agreed that as long as it is technically possible at a reasonable cost, the requirement should not be given up.

5.3 Inner tube

With respect to reproducibility, the inner tube is one of the main limiting parts of the chamber. There is a trade-off in defining the radius of the tube: a smaller radius results in a higher solid angle and an increased efficiency for low energies while a higher radius weakens the requirements on the tolerance on ampoule positioning.

It was agreed at the BqWG(II) meetings that plastics and composite materials shall not be used in the chamber design, since the effect of radiation on the mechanical and physical properties are (still) unknown. As a result, only a limited number of light but strong metals are suitable.

When made from pure aluminium, the reproducibility requirement translates in a tolerance for the thickness of the inner tube of 1 µm (for 20 keV photons). For pure beryllium and 20 keV photons the tolerance is 24 µm. The use of magnesium alloys was studied as well, yielding in a tolerance requirement of 5 µm.

Tolerance on thickness can in first instance be analytically calculated with the photon linear attenuation formula, but electrons scattered out of the inner tube by higher energy photons will have an important contribution to the signal when the region after the inner tube is part of the sensitive volume. For this reason, calculations should be validated by means of Monte-Carlo simulations and experiments. Discrepancies were found between Geant4 simulations resulting in a tolerance larger than 100 µm and simple transmission calculations, resulting in 24 µm tolerance for the thickness of the beryllium inner tube.

The most promising material for the inner tube is beryllium, but has an extremely high cost of about 50 k€ for a single tube. In addition, beryllium is toxic and listed as a dual-use material which this could be an issue for the deployment of the instrument [Council regulation EC 428/2009].

A beryllium inner tube can be produced with a thickness of 2.5 mm ± 20 µm and an inner diameter of 20 mm. This tube would withstand 2 MPa of pressure, not taking into account the longitudinal stress on the tube caused by the slight bending of the top and bottom lid of the chamber, where the tube is fixed to.

Beryllium grades S-200-F, I-70-H and O-30-H (company Materion) can be electron beam welded or RF brazed to stainless steel 316L (the material for the pressure vessel). Electron beam welding uses a transition material containing aluminium, which is explosively bonded to aluminium for joining to the beryllium side and to stainless steel 316L for joining at the other end. (The cost mentioned above does not include this exotic procedure.)

The tolerances on the composition and density of commercially available metals are of great concern. The material supplier specifies ranges for density and impurities. Based on the specified ranges, the two extremes of the transmission (highest and lowest) were calculated and compared for two beryllium types (Figure 4).

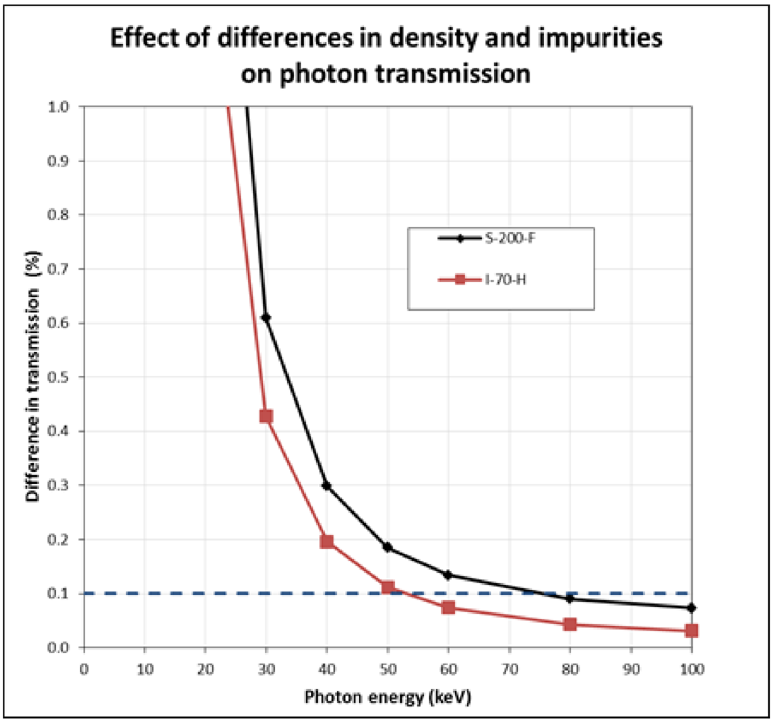

Figure 4 Effect of density and impurities of 2 types of beryllium

The figure shows that the production spread of S-200-F beryllium does not allow reproducing the transmission within 0.1 % for photons with energy lower than about 75 keV, and 0.2 % for energies below about 48 keV. For the more pure type of beryllium, I-70-H (optical grade), 0.1 % reproducibility cannot be achieved below about 55 keV and 0.2 % below 40 keV. These calculations are based on the attenuation formula, and should be verified with Monte-Carlo simulations.

As an alternative of machining a tube, the purchase of a drawn tube (according to specification) was considered, but no company was found capable of achieving the required tolerance. The pragmatic idea to purchase e.g. one kilometre of tubes from a single batch, enough to provide inner tubes for all chambers that will ever be built, was proposed at a BqWG(II) meeting, but quickly rejected, as it would breach the primal objective of reproducibility. Additive manufacturing was investigated as well, but the required accuracy and straightness cannot be guaranteed [BqWG(II) 13/05/2013].

Other ideas were investigated as well, for example the option to create "windows" in the inner tube from a less dense material, but this is very complicated to realise. In addition, ampoule positioning with respect to the windows is likely to become more critical, and difficult to assess. Another idea was to produce the inner tube out of a sheet that is rolled and welded in a tube, but tolerances on sheets are typically one order of magnitude greater than required. In addition, the density might be compromised by the rolling process.

An inner tube out of three sections was considered (Figure 5), where only the middle is highly precise. From theory this is a nice solution, but likely, deformation during welding will make it impossible to properly align the pieces to the required tolerance.

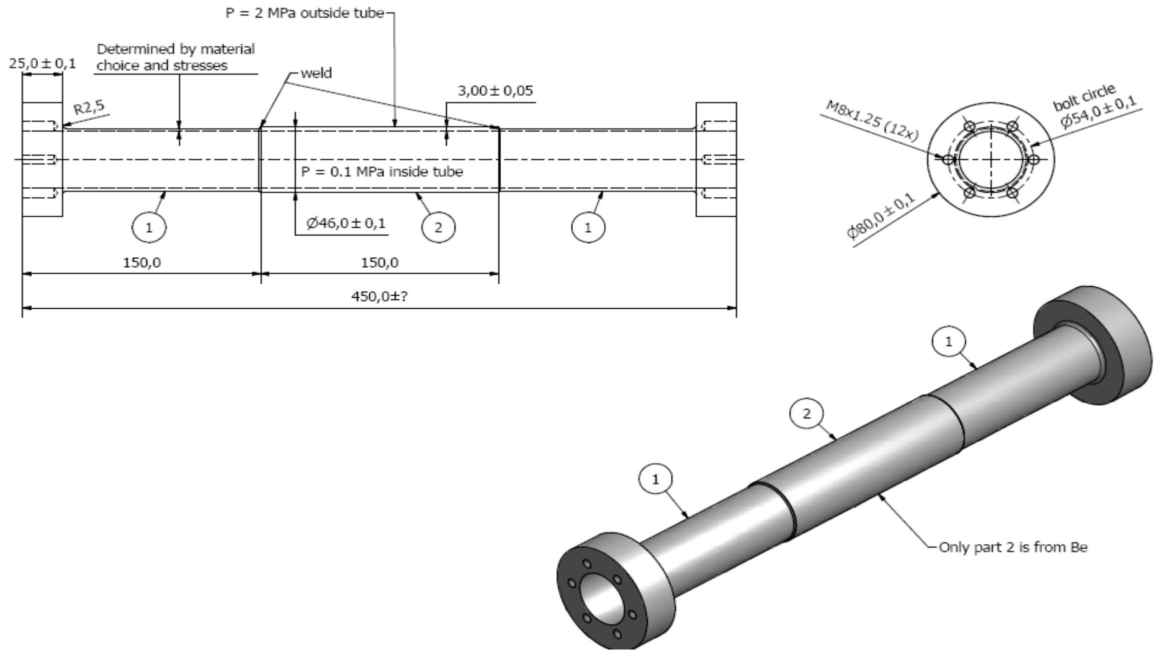

Figure 5: Inner tube from 3 sections

5.4 Gas type and pressure

For the filling gas, nitrogen and argon were considered. Argon yields a much higher response than nitrogen, especially in the energy region between 20 keV and 200 keV[7].

Refilling the chamber with a Ruska 2465A piston pressure gauge yields a reproducible response below the 0.05 % level, measured with a ^{226}Ra source. The pressure gauge generates pressures up to 3 MPa with an accuracy of 0.0035 % of the reading or 1.4 Pa, whichever is greater.

It was decided to use pure argon at a pressure of 2 MPa [BqWG(II) 12/2007].

5.5 Bias voltage

The bias should be high enough to reach linearity between the measured current and the activity. A voltage of 1000 V is not enough[7] to limit recombination effects when strong sources (> 100 MBq) are used.

5.6 Ampoules

The radioactive solutions are measured in glass ampoules with a volume of 5 mL. Changes in the composition of the glass and the thickness of the wall of the ampoules affect the absorption of gamma rays, especially below 50 keV.

Camps [BqWG(II), 22/05/2007] reports results from Monte-Carlo simulations:

- The effect of the wall thickness is larger than the effect of irregularities of the bottom of the ampoule, which only are relevant below 40 keV, as reported also later by Amoit [BqWG(II) 02/11/2010];
- The ampoules are reproducible up to 0.1 % for gamma energies above 50 keV;

- Tests have shown that the SIR ampoules are not reproducible for low energy. Differences up to 1.2 % at 20 keV were observed due to large differences of wall thickness.

The ampoules used by the SIR are from a single batch. Although the quantity available is limited, BIPM estimates to have enough SIR ampoules up to 2060.

Suliman showed that the SIR ampoules have smaller variations than the IRMM ampoules, which are from another batch. Figure 6 shows measurements of the wall thickness, performed with an Olympus Magna-Mike 8500 Hall-effect thickness gage. During the measurements, the ampoule was rotated manually. Readings are plotted versus time. The IRMM ampoule made 8 revolutions, as can be observed on the graph. Such measurements were performed at different vertical positions in the ampoule, leading to similar results.

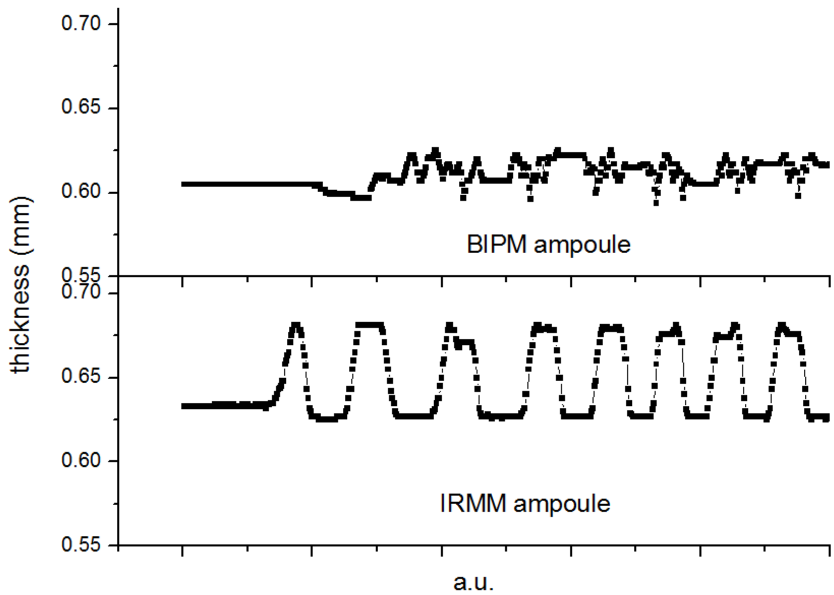

Figure 6: Variations in the thickness of the wall of SIR/BIPM ampoules and IRMM ampoules.

Until now, no characterisation of the upper part of the ampoule has been performed.

In principle, all SIR ampoules are filled with 3.6 (2) g of solution. There is an observable effect of the filling height of the ampoules. It was discussed that the source holder should allow a vertical centring of the solution, to minimise and simplify corrections that have to be applied. Such a source holder introduces the risk of unintentional movements of the ampoule and consequently measurements at a wrong position. A compromise is to make one fixed holder, and one which allows vertical movement of the source, clearly distinguishable from each other.

5.7 Source holder

The ampoules are placed in a source holder, such as the one depicted in Figure 7. The source holder should ensure that the ampoule is always placed in the same position with respect to the sensitive volume of the chamber. A cone-shape connection between the holder and inner tube improves the reproducibility of source positioning. Care should be taken with modular source holders consisting of different parts, as the parts may shift over time, causing a difference in response. For the same reason, the use of glue is not recommended as it may come loose.

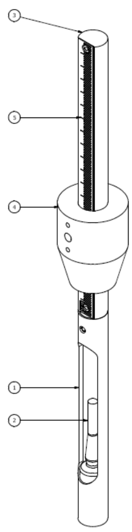

Figure 7: Source holder

To achieve a reproducibility of 0.2 % at 20 keV, the tolerance on the radial source positioning shall be:

- 0.9 mm for 20 keV photons in 40 mm diameter well (Camps and Paepen, [7])
- 0.5 mm for 20 keV photons in 20 mm diameter well (Suliman)

Several sub-parts of the source holder were produced at the workshop of IRMM and one of each sub-part at the workshop of BIPM. To test the reproducibility, these parts were interchanged in all possible combinations, but keeping the stick and conical ring always together. Previous tests showed indeed that only the lower part of the holder (with the ampoule) is critical. On a prototype chamber of IRMM, the ionisation current of a 70 MBq ^{109}Cd source was measured in all the source holder configurations.

One concludes that:

- Workshops have difficulties in producing a reproducible bottom part of the holder. Measurements of the wall thickness showed that the BIPM holder has the most uniform wall thickness and that IRMM holders no not have a uniform wall thickness at all.
- The magnetic thickness gauge is not accurate enough for absolute thickness measurements. It measures the disturbance of the magnetic field between a steel ball and the probe (a permanent magnet). The manufacturer likely understated the claimed measurement uncertainty (1 % + 3 μm), or other effects have an important effect (temperature, calibration, …). As such, the gauge can only be used to give a relative trend.
- The lower part is currently not strong enough, as one item broke during the tests.
- With conventional callipers, the critical bottom part of the holder cannot be reached for thickness measurements. Acceptance tests are difficult with this holder design.

The design of the holder should be reconsidered. Additive manufacturing could provide a solution: a holder with minimal amounts of material, yet strong enough may be 3D printed. When light plastics and a minimal amount of material are used, the tolerance becomes less strict for low energy photons. Metals may also provide a solution, since they can be 3D printed with a higher precision.

6 Traceability

6.1 Requirement

To ensure reproducibility, the values and tolerances of all the parameters influencing the response must first be unequivocally specified in SI units wherever possible and they must be capable of being measured in a manner which ensures that the actual values of the realised parameters are traceable to the SI. Also the ionisation current generated by the chamber should be measured in a manner which ensures full traceability to the SI, over the full range required, from a few tens of fA (for background measurements) up to hundreds of nA.

6.2 Measurement of the ionisation current

The measurement of the electric current generated by the chamber is performed by an electrometer. An example of such a device is the Keithley 6517 electrometer. These instruments integrate the charge over a capacitor placed in the feedback loop of an operational amplifier. Input offset currents of the operational amplifier should be as low as possible. Commercially available electrometers have a set of built-in feedback capacitors, which have to be reasonably small to be able to fit inside the device's housing. This limitation compromises the quality of these capacitors. The result is that the calibration of the commercial current meters is not sufficiently accurate.

The preferred way of measuring the current is by operating the electrometer with a separate, high-quality capacitor placed in the external feedback loop of the device. in principle, if the capacitance is precisely known and sufficiently large compared to the stray capacitance of the cables and connections to the electrometer, no calibration of the current measuring system as a whole is required.

The insulation resistance between the two terminals of the capacitor, and to the housing, shall be larger than 10^{16} Ohm, which corresponds to a leakage current of 1 fA at 10 V. The leakage current or insulation resistance is of key importance, but not always specified or guaranteed by the manufacturer. This is due to the fact that standard capacitors are mainly used for the calibration of LCR meters, where the leakage current is not as important as in this application. The leakage currents of commercially available capacitors as well as capacitors built in-house were measured when biased to 20 V, resulting in leakage currents ranging from 4 fA to about 5 pA.

An issue with capacitors is that they are calibrated by LCR calibrators, using an alternating current with a frequency of 1 kHz. In this application, they are charged with a direct current. Research by Rietveld and van den Brom[9] showed that there is a difference between the AC and DC capacitance when the medium between the plates is not dry. For that reason, only hermitically sealed, inert gas filled capacitors are suitable. In any case, the quality of such capacitors should be followed up.

A charged capacitor is itself an ionisation chamber. Capacitors should be placed in lead shield to avoid discharge by natural background radiation when they are charged to about 10 V. Also, the decay of radon penetrating the capacitor housing may discharge it, which is another reason to use sealed capacitors.

The effect of stray capacitance and the difficulties with the calibration of standard capacitors can be controlled by calibrating and regularly verifying the current measuring system as a whole (the electrometer, external feedback capacitor and cables). The most precise way of generating an SI traceable DC current is by using a voltage ramp generator and a standard capacitor, as demonstrated by van den Brom et al.[10] This technique is available at a few NMIs and also IRMM has built such a current source.

To conclude, the measurement of the ionisation current is difficult and requires expertise, but is feasible.

7 Stability

7.1 Requirement

The response curve of the chamber (generated current per becquerel as a function of gamma energy) shall be stable over tens of years.

7.2 Discussion

Gas leaks are a main concern with respect to long-term stability. Preferably, the pressure vessel should be welded. However, when different materials are used, e.g. stainless steel and beryllium, welding becomes an issue. In addition, thermal deformation due to welding should be under control. Laser or electron beam welding could provide a solution to the latter.

The electric feedthroughs should be extremely reliable and tight. A commercially available solution was found and may be provided by a supplier of connectors for deep underwater applications (undersea cables). Such feedthrough consist of two glass seals, specified with an insulation resistance of at least 1 GΩ at 500 VDC, pin to shield, shield to body and pin to body. (There is no potential difference between the contacts. The inner one carries the current to be measured.) The seals are rated to withstand 69 MPa in both directions. The connector has a 9/16-18 SAE port, which can be screwed into the chamber and sealed with an o-ring (during R&D). The body can also be electron beam welded to the chamber, provided that the temperature of the glass does not exceed 120 °C. Even after welding, the feedthrough can be removed and replaced in case of malfunctioning.

The stability of the chamber could be verified by measurements with sources with a long half-life, providing that these do not leak. Alternatively, pressure and temperature sensors may be used to assess the long term stability. However, such sensors are not stable and precise enough on the long term, requiring regular recalibration. Recalibrating a sensor means that it has to be removed from the chamber, which poses additional problems since it is unavoidable that a small volume of gas is lost in that process.

The thermal expansion of the chamber affects the response. Also the current measuring system is affected by temperature. The chamber should be operated in stable environmental conditions, which should be feasible.

8 Stress analysis

8.1 Assumptions

Stress analysis by finite element analysis was performed on the 2010 design, with 2 MPa of gas pressure. To reduce production cost, flanges were removed and local reduction of the outer radius was proposed for the bolts (Figure 8).

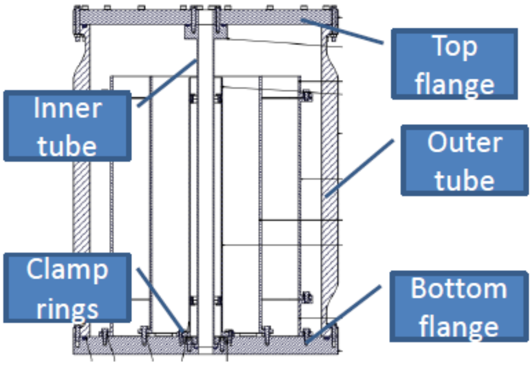

Figure 8: The 2010 design without flanges, for the first stress analysis

To reduce the complexity of the calculations, following simplifications were applied:

- O-ring grooves were discarded to reduce the number of mesh elements;
- Non-load carrying electrodes were removed, as this has no influence on system behaviour;
- Unused holes were removed;
- Bolts were removed and replaced by virtual bolts (spring features with same properties as the section of the real bolt), and a pre-load of 90% of the yield limit was applied;

For the stress analysis, the outer tube, top and bottom flanges are from 316L stainless steel, the inner tube from beryllium, type I-70-H.

8.2 Results

The gas pressure causes top and bottom flanges to bend outward (Figure 9). Note that the deformation in all the graphs is scaled by a factor of 30. At the centre, the flanges move apart with 1.05 mm. The global stresses are well below 300 MPa (the assumed yield limit for stainless steel).

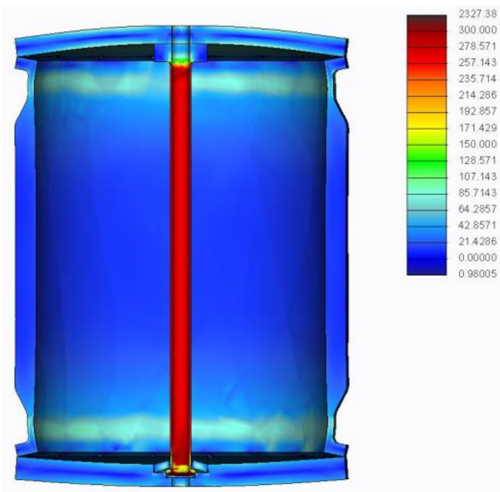

Figure 9: Deformation (scaled 30x) and stress (MPa)

Local stresses at bolt contacts are slightly below the yield limit (Figure 10).

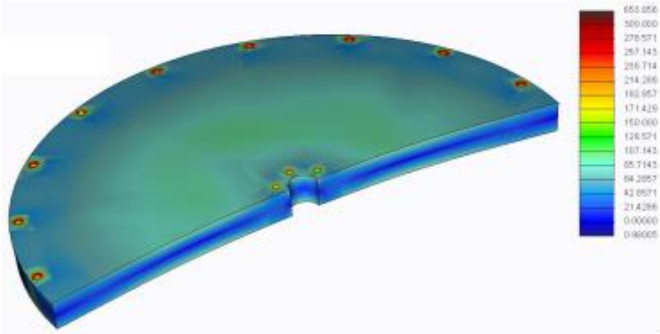

Figure 10: Local stress at bolt contacts

The thin regions in the outer tube cause the tube flanges to bend inwards. Local stresses slightly exceed yield limit of 200 MPa for aluminium, but are less than the yield limit for stainless steel (Figure 11).

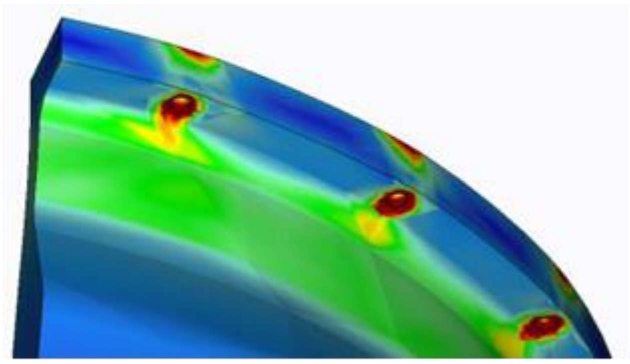

Figure 11: Local stress at outer tube flanges

Both previous effects cause excessive gaps of 0.16 mm at the place where the grooves for the o-rings are, resulting in insufficient squeeze and high probability for leaks or extrusion (Figure 12, not showing the groove itself).

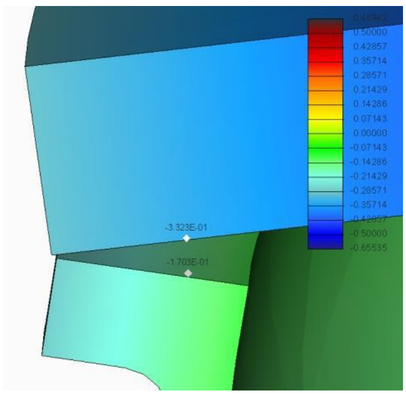

Figure 12: Bending of top flange and outer tube result in risk for o-ring extrusion.

The inner tube is stretched (Figure 13). Stress over the majority of the length of the tube is 273 MPa, due to tensile load caused by the outward movement of the flanges and the pressure. Controlling the bending of the flanges better will reduce the stress below the maximum for Be I-70-H (207 MPa). At the top of the tube, a round is advised to decrease local stresses.

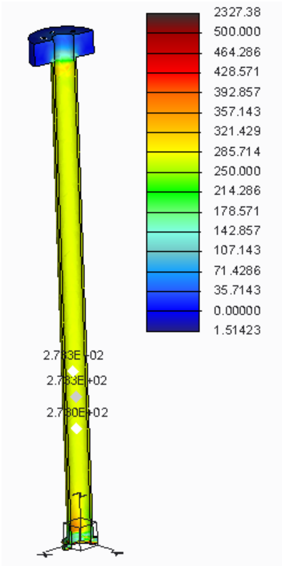

Figure 13: Stretching of the inner tube

At the bottom of inner tube, stresses exceed 1000 MPa, which is problematic (Figure 14).

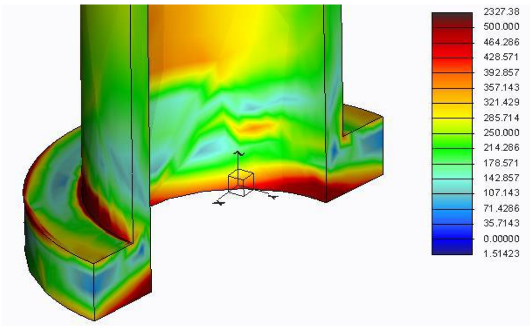

Figure 14: Excessive stress is observed at the ends of the inner tube

The two clamp rings at the bottom of the inner tube tilt open (Figure 15). Stress reaches 700 MPa, much higher than yield strength of 300 MPa. Local stresses under bolt heads also exceed yield limit.

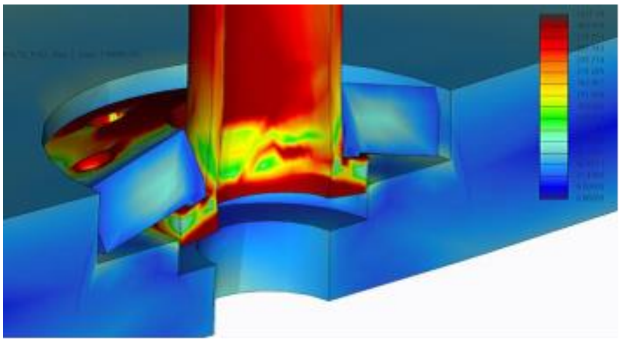

Figure 15: Excessive stress in the clamps

8.3 Recommendations

Based on the results of the stress calculation, the following changes are recommended:

- Aluminium is not strong enough for the outer tube. Stainless steel 316L should be used;
- The outer tube should have no flanges but threaded holes. There should be no thinning at the ends, to increase strength;
- The top flange should be from stainless steel 316L, have an increased thickness to 30 mm to reduce bending and longitudinal stress on inner tube, and should have 24 bolt holes. M8 bolts, stainless steel, ISO 898/1 class 12.9 shall be used;
- The bottom flange is should be from stainless steel 316L, contain 24 bolt holes and have a thickness of 35 mm to reduce bending and longitudinal stress on inner tube. M8 bolts, stainless steel, ISO 898/1 class 12.9 shall be used. The bolts shall be recessed so that the chamber does not rest on the bolt head;
- The inner tube may be made from beryllium type I-70-H. The edges at the top and bottom shall be rounded to reduce local stress. M5 or M6 bolts shall be used;
- During prototyping and testing, when the chamber is opened and closed many times, o-rings may be used. A metal-to-metal contact needs to be assured to control the compression of the rings and to prevent extrusion of the o-ring. The design should be such so that later welding is still possible.

Stress analysis is an essential part of the design process, to ensure reproducibility and safe operation of the instrument.

9 Response

9.1 Linearity requirement

Within a certain operating range, there shall be a linear relation between the generated current versus the activity, for the gamma energies from 30 keV to 2.6 MeV.

9.2 Charge collection – electrode design

An important improvement applied in the 2010 design is the decision not to use the electrodes as structural elements to hold the gas pressure. The sensitive volume of the chamber is now much better defined and easier to reproduce. To study the charge collection, the electric field was calculated by finite element analysis obtained with ELMER. The electrical field lines were used to predict the place on the electrodes where electrons and ions would be collected, not taking into account recombination and diffusion. The location of interactions in the gas was obtained from Geant4. This approach results in a first approximation of the sensitive volume.

The following conclusions were made (applicable to the 2010 design):

- The collection of charges matters significantly depending on the design of the electrodes. In order to estimate the response of the prototype, a simulation (or any other type of calculation) is essential and shall include a way to describe the areas from which the charge is collected, e.g. by applying the Shockley-Ramo theorem, and including diffusion and recombination effects.

- The first (inner) electrode collects always more charge than the second. This unequal distribution of the current between the two electrodes should be taken into account when the electric circuits for reading the chamber signal are designed.

- The insulators separating the guard rings seem appropriate, but as a design goal, they should be as small as possible. An additional observation is that for the purpose of defining the collection volume, the insulators on the outer ring are more important than those on the inner ring. An overlapping structure, as described in Figure 16, is recommended[8], to reduce the volume of uncertain charge collection.

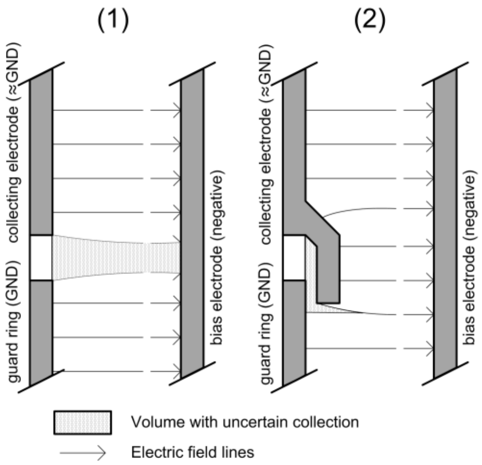

Figure 16: Overlapping structure to reduce volume with uncertain charge collection

- Of high interest is the volume between the inner tube and the first collecting electrode, where a significant charge is deposited (Figure 17). The maps show that, of the charge deposited in the gas, the bulk is deposited between the collecting electrodes.

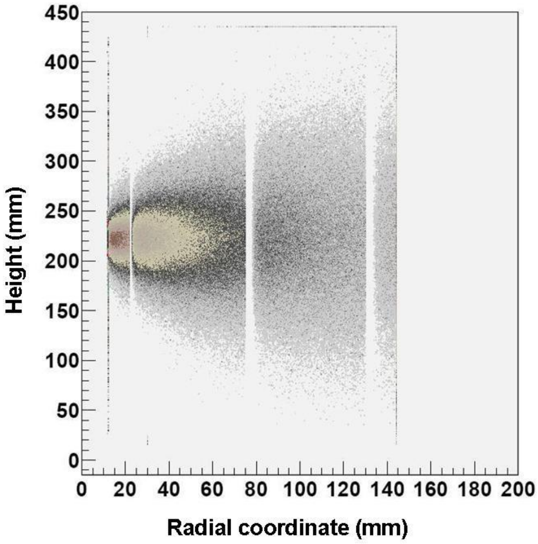

Figure 17: Deposited charge map for 1 MeV photons

- If the inner tube is made from aluminium type 2024, almost no charge is collected from 20 keV photons. Replacement with beryllium leads to a significant improvement.

10 Extension to pure beta emitters

Beta radiation interacts differently than gamma radiation, and this affects the response of the chamber. It is a wish to be able to extend the use of the chamber to pure beta emitters, but this is difficult to realise. Amiot [BqWG(II) 09/05/2012] reported a comparison between the response derived experimentally and obtained by calculations: large relative discrepancies were observed (about 30 to 80 % for commonly used pure beta emitters). The use of a liner to stop betas and convert them to bremsstrahlung photons before they enter the chamber has been investigated.

11 Conclusion

This report starts with the question whether it is feasible to realise a distributed radioactivity standard; an instrument that generates a reproducible response when constructed according to a full set of design specifications and tolerances.

After many years of research, prototyping and testing, the answer to that question is still indecisive. Many issues have been addressed and resolved, but additional issues have been raised as well. The requirement on the reproducibility at low energy is tremendously demanding. Not giving up this requirement implies the use of the extremely expensive, toxic and dual-use material beryllium. And yet, even for the purest form of beryllium, the variation in concentrations of impurities between production batches result in a breach of the reproducibility requirement at low energy. Allowing a less strict reproducibility at lower energy would enable the use of aluminium or magnesium alloys for the construction of the inner tube, but still the requirements on tolerances of the thickness would be challenging.

The conclusion of this work is that even if it would be physically and technically achievable to build such an instrument, the realisation of a distributed radioactivity standard will be far too expensive and complicated for National Metrology Institutes.

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List of abbreviations and definitions

BIPM	Bureau International des Poids et Mesures (International Bureau of Weights and Measures)
CCRI	Consultative Committee for Ionizing Radiation
IRMM	Institute for Reference Materials and Measurements (JRG Geel since 1 July 2016)
NMI	National Metrology Institute
SI	Système international d'unités (international system of units)
SIR	Système International de Référence (International Reference System)
SIRTI	Système International de Référence Transfer Instrument

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