THE NEXT GENERATION OF CURRENT MEASUREMENT FOR IONIZATION CHAMBERS

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Abstract

Re-entrant ionization chambers (ICs) are essential to radionuclide metrology and nuclear medicine for maintaining standards and measuring half-lives. The requirements of top-level metrology demand that systems must be precise and stable to 0.1% over many years, and linear from 10^{-14} A to 10^{-8} A. Thus, laboratories depend on bespoke current measurement systems and often rely on sealed sources to generate reference currents. To maintain and improve present capabilities, metrologists need to overcome two looming challenges: ageing electronics and decreasing availability of sealed sources. Possible solutions using Ultrastable Low-Noise Current Amplifiers (ULCAs), resistive-feedback electrometers, and (quantum) single-electron pumps are reviewed. Broader discussions of IC design and methodology are discussed. ULCAs show promise and resistive-feedback systems which take advantage of standard resistor calibrations offer an alternative.

1 Introduction

The science of radionuclide metrology relies heavily on pressurized reentrant ionization chambers (ICs), from national standards through to nuclear medicine clinics – in fact, the origins of such instruments can be traced back to the discovery of radioactivity (Thomson and Rutherford, 1896). Nowadays, at the international level, the International Reference System (SIR) (Rytz, 1983) relies on a pair of ICs used to compare national standards of radioactivity so that countries can confirm equivalence of their standards. Most national metrology institutes (NMIs) maintain a set of ICs that act as the 'memory' of primary standards of radioactivity; essentially secondary standards, the instruments are reproducible and extremely stable over several decades, so it is possible to use them to replace standardized solutions, avoiding the need to repeat complex realizations of primary standards. The stability of ICs means that they are also ideal for clinical use, checking the activity content of radiopharmaceuticals before administration to patients for diagnostic scans or radionuclide therapy (Zimmerman and Judge, 2007).

One challenge of using such instruments is the wide range of electrical currents that must be measured. The response (in pA) of the instrument depends on the activity (in Bq) of the artefact being measured, and on the intensity and energies of the gamma-rays emitted in the decay of the radionuclide. The activity to be measured can range from GBq for medical applications down to kBq for replicating standardized solutions, and the energy range to cover is typically from 60 keV (for ^{241}Am standards) to 2000 keV or higher (for radionuclides such as ^{88}Y). The range of electrical current to be measured is therefore wide – typically 10^{-13} A to 10^{-8} A – and with no sample in the chamber, the background current is typically 10^{-15} A (Schrader, 1997).

The precision and stability needed are also challenging. Primary standards of radioactivity can be realized with uncertainties of 0.15 % to 0.5 % (depending on the radionuclide), so repeatability

and stability of better than 0.1 % are needed to compare national standards or to replicate the primary standards. Clinical applications of such instruments are less demanding – Gadd et al., (2006) state the repeatability and linearity should be better than 1 % and 5 %, respectively.

For half-life measurements, ionization chambers are nearly ideal since they do not suffer dead-time and can measure radioactive decay over multiple half-lives. Variability in current offsets over days or weeks can be the dominant source of uncertainty (Pommé, et al., 2008). Under ideal conditions, root-mean-square fit residuals as low as about 0.02 % from multiple current readings over a few days have been achieved (c.f. Schrader 2004 Fig. 4). Commercial measurement systems often show range change (Schrader 2004 Fig. 3) and cyclic environmental effects (Schrader 2016) at least 20 times larger, which can dominate the uncertainty in the fitted half-life and even be confused with changing physics (Pommé et al, 2017).

Ionization chambers therefore require electrical current measurements with high precision (better than 0.1 %) and linearity over a wide range (more than 5 orders of magnitude). Investigations of the uncertainty budget for ionization chamber measurements (Amiot et al., 2015) have shown that the linearity is one of the major components of the uncertainty. The linearity problem has led many institutes such as the BIPM (Rytz, 1983) to use a set of long-lived sealed radioactive sources to produce reference currents against which the current produced by the standards may be measured – the measurement becomes a ratio of currents, close to unity and therefore more reproducible. Although this solves the linearity problem, eventually the safety of retaining sealed sources due to radiation damage to and pressure build-up inside the source capsule can be called into question.

A strong motivation for improving current measurement is to identify physical issues with these ageing measurement systems before they cause serious measurement errors. For example, both

the pressure in the chamber and position of the sample would not change the ratio of currents from two reference ^{226}Ra sources but would cause errors in the resultant activity for another nuclide based on measurement relative to a ^{226}Ra reference source. Such an issue occurred at NIST, where a slow drift of the source position over 40 years caused systematic errors in half-life and activity measurements well beyond claimed uncertainties (Fitzgerald, 2012). A contributing factor to the delayed discovery of this problem was that the precision electrometer relied on an air capacitor, which was adequate for ratios of similar currents from a reference source but had large (1 %) seasonal variability which masked the drift in absolute current.

There have been many advances in low electrical current measurement since ionization chambers became an established part of the international radionuclide metrology infrastructure. This paper, which is rooted in a joint NIST-BIPM workshop held in September 2018, sets out to address the question of whether these technological advances could offer better precision and accuracy for ionization chamber measurements, reducing the dependence on sealed reference sources and improving further the long-term viability of the technique.

These motivations have drawn increasing interest from the radionuclide metrology community. Following the 2018 workshop attended by representatives of 8 NMIs and the BIPM, in 2019 the Consultative Committee for Ionizing Radiation (CCRI) and Consultative Committee for Electricity and Magnetism (CCEM) started a joint Task Group on this topic (CCRI-CCEM, 2020).

In Section 2 and Section 3 we will summarize the importance and operating principles of ICs. This is followed by a description of traditional (Section 4) and new (Section 5) approaches to IC current measurement, and finally a summary and outlook in Section 6.

2 The importance of ionization chambers

The first step in using an ionization chamber at an NMI is to realize a primary standard of the radionuclide of interest. The ionization chamber can then be calibrated for that radionuclide (*N*), by measuring a source with activity (A) that produces current (I), resulting in a response factor $\varepsilon_N = I/A$.

Alternatively, a response ratio (*K*, sometimes called 'equivalent activity'), relating the ratio (*R*) of currents measured for the primary standard to that from a sealed reference source, can be defined as $K = A/R$. The ratio method reduces some uncertainties (e.g., due to temperature and humidity effects) and is therefore often preferred for measurements over long timescales.

The main application of ICs at NMIs is replicating primary standards of radioactivity. However, researchers often rely on ICs to benchmark their activity standards against other laboratories (e.g., Ratel 2007; Woods and Baker, 2004) and against previous results at their own institutions (e.g., Bergeron et al., 2014). For either application, it is critical that the IC calibration adds negligible measurement uncertainty so that the primary activity standards, with their attendant uncertainties, are the true comparands. The long-term stability afforded by ICs has been leveraged to provide some of the most precise determinations of the half-life of many photon-emitting radionuclides (e.g., Schrader, 2004).

Beyond the radionuclide metrology community, ICs (often referred to as 'dose calibrators') are the principal means of measuring radioactivity in nuclear medicine clinics. NMIs often work with device or radiopharmaceutical manufacturers to determine the appropriate calibration.

Settings determined at NMIs are published (e.g., Bergeron and Cessna, 2018) but users are encouraged to determine their own device- and geometry-specific settings for a given nuclide. Calibration protocols involving long-lived surrogates for short-lived radiopharmaceuticals have emerged as an approach to achieving traceability to national standards (Zimmerman and Cessna, 2010).

3 Ionization Chamber Operational Principles

The operation of a well-type ionization chamber is shown in Figure 1. The radioactive material (usually a solution sealed in a glass ampoule) is placed near the center of the well in the instrument. The chamber is hermetically sealed and contains a gas at high pressure (typically 1 MPa of nitrogen). Gamma rays emitted in the decay of the radionuclide ionize the gas in the chamber, then the freed charge is collected by applying a high voltage (typically 500 V) across the volume of the chamber. The current produced is proportional to the activity of the sample; once calibrated in terms of pA/MBq using a primary standard of radioactivity, the measurement can be used to determine the activity content (MBq) of the ampoule. Schrader (1997) has published a comprehensive review of the operation of an ionization chamber.

Numerous techniques have been used to measure the electrical current. The current produced in the ionization chamber is almost constant during a measurement, other than small fluctuations due to the statistics of the decay and detection processes, electronic noise and variations in background radiation (Schrader, 1997). This means that quasi-static methods such as charge integration with capacitors are commonly used.

The linearity of the measurement over a wide range of currents is an important parameter – for applications in metrology institutes, a linearity of better than 0.05 % over 5 orders of magnitude of current is required. Non-linearities can be minimized by selecting the capacitance or voltage such that the electrometer is always used in the same small range (Weiss, 1973). The linearity can be checked using an accurate current source or by following the decay of a sample of a short-lived radionuclide (such as ^{18}F). More than one value of capacitor may be required, and non-linearities in current versus activity have been observed when switching from one capacitor to another (Schrader, 2004). At very high ionization rates in the chamber, the linear relation between activity and current is lost due to charge recombination effects (Schrader, 1997).

Two further parameters of importance are the stability and reproducibility of the measurements. Reproducibility may be limited by changes in the temperature in the laboratory and variations in the background current – these can be overcome by accurate control of the room temperature and ensuring short measurement times (typically less than one hour). Long-term instabilities may be due to drifts in the electronics, slow gas leaks reducing the chamber pressure and changes in the sample geometry. Such instabilities can be monitored by checking the electrometer with an accurate current source or measuring a reference source with a long half life, such as ^{226}Ra (Mann, 1958; Unterweger and Fitzgerald, 2012).

The issues of linearity and stability have been addressed at the BIPM and NMIs by measuring the ratio of the current produced by the source to the current produced by one of a set of sealed reference sources. This is supported by measurements at the BIPM which have shown that the

ratio of currents produced by two ^{226}Ra sources has been stable to around 0.04 % since 1976 (

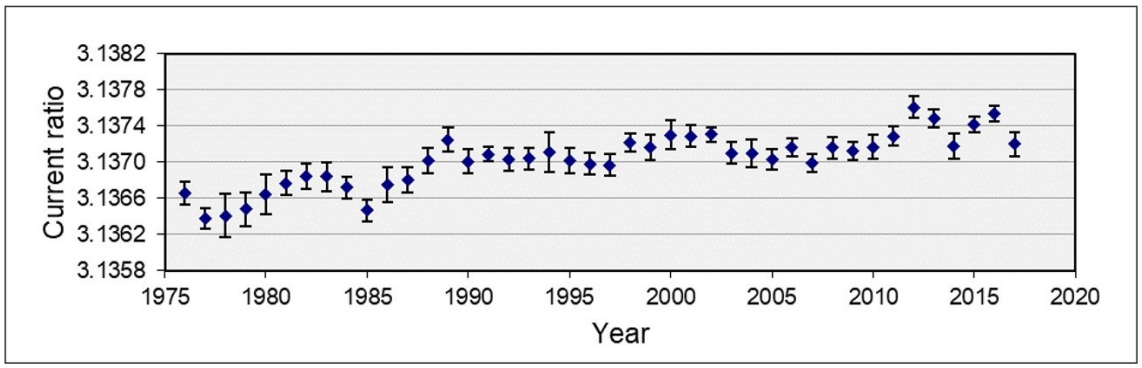

Figure 2) while the measured current produced by a single reference source has been stable only to 0.1 %. However, a long-term trend in the ratio can be observed− origin of this effect is unknown. Exacerbating the problem, it is likely that many laboratories will no longer be able to keep usual electrometers or reference sources for as long as 40 years ("forever"), making it even more important to have highly stable IC current measurement systems linked to the SI.

Other factors contributing to the overall measurement uncertainty are related to the item being measured (for example, the thickness of the glass walls of an ampoule, the filling height and the chemical form / density). Corrections also must be made for decay during measurement and the effect of gamma-ray emitting impurities.

4 Conventional current measurement systems

The measurement of DC electrical currents below 10 nA is possible with two types of instrument: the integrating electrometer and the feedback ammeter. The principle of the electrometer and ammeter are illustrated in Figure 3. Both use a high-gain amplifier with a feedback element. The feedback element for an electrometer is a capacitor (capacitance, C), so that an input current yields a time-dependent voltage V_{OUT} (t) at the output of the amplifier

according to $I = C\ \mathrm{d}V_{\mathrm{OUT}}/\mathrm{d}t$. In the case of the ammeter, the feedback element is a resistor, yielding a constant output voltage according to $I = V_{\mathrm{OUT}}/R$.

In the next two sub-sections we briefly review the advantages and limitations of each type of instrument.

4.1 Integrating electrometer

Integrating electrometers are often used with ICs at NMIs. The amplifier, feedback capacitor and voltmeter are separate units, with the amplifiers often being custom-made. Currents in the range of 1 pA to 1 nA require capacitors in the range 1 pF to 1 nF to give reasonable voltage ramp rates of around 0.1 V/s to 1 V/s. Low-loss sealed-gas-dielectric standards are readily available for this range: such capacitors can have phase angles less than 10^{-5}, resulting in a very small residual non-linearity to the voltage ramp, and have low relative sensitivity to temperature – a few 10^{-6}/°C is typical. If a sealed-gas-dielectric capacitor is used, there is no dependence on humidity. Unsealed air-dielectric capacitors in an uncontrolled environment will exhibit variations in capacitance with humidity of up to 0.05 % (Ford, 1948). Traceable calibrations of capacitance, voltage, and time with relative uncertainties in the range of 10^{-6} or 10^{-5} are available from NMIs and commercial calibration laboratories.

In theory, an integrating electrometer should be able to measure current with a relative uncertainty in the range of 10^{-5}. In practice, the uncertainty is typically in the range of 10^{-4}, an order of magnitude higher than the combined uncertainties in the calibrations of capacitance, voltage, and time. The two main additional components are frequency dependence in the capacitor (Giblin et al., 2010), and the presence of stray capacitance in the amplifier, in parallel with the feedback capacitor (Giblin et al., 2009; Giblin and Lorusso, 2019). It is worth

emphasizing the frequency dependence effect. The uncertainty in the capacitance calibration, which is typically performed at a frequency of 1 kHz, cannot be interpreted as the uncertainty in the capacitance when used in a ramp electrometer at a frequency much less than 1 Hz. (Giblin et al., 2010). The stray capacitance results in a larger non-linearity to the voltage ramp than would be expected from the properties of the feedback capacitor alone. Despite these limitations, electrometers have proven to be robust and reliable.

4.2 Feedback ammeter

In the last 20 years, specialized low-current ammeters have become commercially available. Some ammeters contain all the functional elements of Figure 3 (b) in one unit and provide a readout in amps. Others are more correctly termed current preamplifiers, as they require a voltmeter to record the output. In either case, the limitation to the achievable uncertainty is the stability and temperature-coefficient of the feedback resistor. These are usually thick-film elements in the range 1 GΩ to 1 TΩ, with relative temperature coefficients in the range 10^{-5}/°C to 10^{-4}/°C and higher sensitivity to mechanical shock than the capacitors discussed in the previous section.

One issue with commercial ammeters is that designers must balance the current noise and voltage noise (Giblin and Lorusso, 2019). Designers normally seek to minimize the current noise; however, ionization chambers have very large output impedances, so this compromise is not needed and custom-designed ammeters can have better performance.

In an IC-readout application, low-current ammeters can achieve comparable performance and uncertainty to an electrometer (Giblin and Lorusso, 2019). Ammeters also have the advantage

over electrometers of providing a continuous measurement, without the need for a charge-discharge cycle.

An integrated one-box ammeter must be calibrated by supplying a reference current to the input. Low-current calibration services with relative uncertainties as low as 10^{-5} are offered by NMIs, but the achievable uncertainty will be limited by long-term (month to year) stability of the instrument. Lower uncertainties may be achievable using commercial ammeters if they are calibrated with a stable on-site reference, such as the ULCA or standard resistor as described in Section 5.

In a study of an electrometer (as described in Section 4.1) and a commercial ammeter using an accurate current source, it was found that the electrometer had a systematic bias of about 0.05 % (Giblin and Lorusso, 2019) whereas the ammeter, following calibration, had no such problem. The problem with the electrometer may be due to the frequency dependence of the integrating capacitor (Giblin et al., 2010), but the cause has not yet been found. This is not a problem for IC measurements if the ratio method is used but could be significant in some cases.

In practice, widely-used commercially-available electrometers (Section 4.1) and ammeters (Section 4.2) are limited in 1 year relative accuracy to about 1 % at 200 pA (and at 2 μC) and 0.2 % at 2 nA (0.4 % at 200 nC), in current or charge mode, respectively (Keithley, 2020). Potential biases are largest at range changes, which can limit uncertainties for specific currents and for half-lives (See Figure 3b of Schrader, 2004). The new technologies presented here (Section 5) can reduce these limiting biases and uncertainties.

5 New technologies

In this section, we will look at three techniques which could change the approach to ionization chamber current measurement – the Ultrastable Low-noise Current Amplifier (ULCA), Single-Electron Pumps (SEPs), and on-site calibration.

5.1 Ultrastable Low-noise Current Amplifier (ULCA)

The Ultrastable Low-noise Current Amplifier (ULCA) (Drung et al., 2015a; 2015b; 2015c) is a novel two-stage transimpedance amplifier with specially-designed operational amplifiers and resistor networks, as shown in Figure 4. The first stage provides a 1000-fold amplification of the input current, and the second stage performs a current-to-voltage conversion via an internal 1 MΩ precision resistor. The transfer coefficient of the ULCA, its transresistance A_{TR}, is nominally $1000 \times 1\ \text{M}\Omega = 1\ \text{G}\Omega$. The current I_{IN} is determined from the output voltage of the ULCA, $U_{OUT} = A_{TR} \cdot I_{IN}$, measured with a calibrated voltmeter.

A_{TR} is calibrated traceable to a quantum Hall resistance with a standard uncertainty of about 14 nΩ/Ω (i.e., relative uncertainty of 1.4 x 10^{-8}) by calibrating the input and output stages in separate steps using a cryogenic current comparator (CCC) (Drung et al., 2015a; 2015b). By using a voltmeter calibrated traceable to a Josephson voltage standard, the input current I_{IN} is measured traceable to the SI values of the von-Klitzing and Josephson constants.

The ULCA in combination with an additional voltage source can also be used as a current source for calibration of electrometers or ammeters (Drung et al., 2015a; 2015c); relative uncertainties of the order of 10^{-6} have been demonstrated (Giblin et al., 2019; Scherer et al., 2018).

The ULCA has shown excellent stability of A_{TR} with respect to time, temperature, current amplitude and transportability. The annual drift of A_{TR} is typically less than 2 μΩ/Ω (Krause et al, 2019) and typical short-term fluctuations within one week are about 0.1 μΩ/Ω.

The temperature coefficient of A_{TR} is very small – a typical value is ≈ 0.2 μΩ/Ω/K – so that under normal laboratory conditions temperature corrections are not necessary. For applications requiring the highest accuracy, the ULCA is equipped with an internal temperature sensor.

For ionization chamber measurements, the ULCA in combination with a state-of-the-art voltmeter can be used as an electrometer (see Figure 4). In the example configuration shown, the ULCA is used in "normal" mode for currents up to ±5 nA, i.e. using the internal 1 MΩ metal-foil resistor (A_{TR} = 1 GΩ).

An external standard resistor with resistance R_{ext} can be used for current-voltage conversion. R_{ext} can be chosen up to 100 MΩ so the overall transresistance A_{TR} will be up to 100 GΩ. Further details on the different operation modes and configurations of the ULCA can be found in Drung et al. (2015a, 2015b, 2017).

The ULCA has a noise level of 2.4 fA/√Hz with a low corner frequency of less than 1 mHz, and fast settling: within 3 s after current switching, the relative deviation from the final output value falls below 0.1 μA/A (Drung et al, 2015a). Tests have been carried out from ±1 fA to ±1 μA to evaluate the uncertainties achievable in practice (Scherer et al., 2019). Above about 10 pA, the uncertainty was limited to about 1 μA/A by the voltmeter. Below about 10 pA, the uncertainty was limited by noise, e.g., to about 1 mA/A at ±10 fA.

Two further variants of the ULCA have been described. A low noise configuration has a noise of 1.6 fA/√Hz (1 mHz < f < 0.5 Hz) (Krause et al, 2019), reducing the measurement time for small

currents by a factor of two. A low-current configuration has an increased $A_{TR} = 5\ G\Omega$ ($1000 \times 5\ M\Omega$), a noise level of 0.9 fA/√Hz at 0.1 Hz, and a calibration uncertainty of about 1 μΩ/Ω.

In summary, ULCA implementations have excellent stability at the 10^{-6} level (under normal laboratory working conditions),very good linearity between 10 fA and 10 nA, input protection circuitry, robustness (insensitivity to mechanical shocks), rapid recovery from over-load, low susceptibility to electrical interference, and can be calibrated traceable to the SI.

5.2 Single-electron pumps

This section discusses the possibility of using single electron pumps (cryogenic nanoelectronic devices that generate a current by counting electrons one-by-one) to calibrate the electrometer or ammeter associated with an IC.

Single electron pumps (SEPs) can produce currents around 100 pA with accuracy of about 0.1 μA/A (Kaneko et al., 2016). The power of calibrating the electrometer with an SEP can be captured with a simple formula: the current from the SEP is simply $I = ef$, where e is the charge of the electron and f is a frequency applied to the chip. A SEP is therefore a fundamental standard and is a realization of the SI ampere (Kaneko et al., 2016). The basic concept is that a nanoelectronic chip in a cryogenic system is connected via a long measurement cable assembly to the electrometer used for the IC current measurement. Table 1 shows the relative uncertainty in the current produced, which is several orders of magnitude better than other calibration techniques for electrometers.

There are several challenges to overcome before this technology could be applied in practice. The SEPs require specialist manufacturing, they operate only at low temperature (less than 1 K), connecting the SEP in the cryostat to the electrometer is difficult, and, at present, SEPs have a maximum current of about 0.1 nA to 0.3 nA so could not cover the range needed.

We can conclude this short section as follows: linking the IC current measurement directly to the SI realization of the ampere is a very powerful idea; however, continued development of single electron pumps will be required before this is feasible.

5.3 Electrometer Calibration using a Standard Resistor

An alternative approach being tested at the NIST is including a stable current source in the ionization chamber circuit, to enable frequent calibration of the electrometer.

The currents are generated with a standard resistor and a voltage source in series, where Ohm's Law defines the calibration current. Four ranges were identified (20 pA, 200 pA, 2 nA and 20 nA) with a goal of 10^{-4} or better expanded relative uncertainty ($k = 2$).The standard resistor was calibrated with traceability to the quantum Hall resistance (QHR); a digital volt meter (DVM), to measure the voltage source, was calibrated with traceability to the Josephson voltage standard (JVS). A 1 GΩ resistor was chosen with a voltage source of 0.02 V to 20 V to generate the required currents to calibrate the electrometer at full scale for each range. The relative standard uncertainty for calibration of the 1 GΩ standard resistor was 5 x 10^{-6} and the relative standard uncertainty for calibration of the DVM was 10 x 10^{-6}. Figure 5 shows the ionization chamber, electrometer (I_D), standard resistor (R_S), voltage source (V_S), DVM, and switch (S_1) for connecting the electrometer to the ion chamber or the electrical calibration system.

The resistance standard has been fabricated and calibrated in the NIST resistance laboratory (Dziuba, et al., 1999). The resistance element has been heat treated and hermetically sealed for long-term stability and characterized for temperature and voltage dependence. The drift rate has also been determined to be less than 1 x 10^{-5}/a so the calibration interval for the resistor can be greater than one year.

The voltage burden of the electrometer pushes uncertainty estimates above the target uncertainty at the lower current ranges of 20 pA and 200 pA (Callegaro et al, 2014; Keithley 2016). The voltage burden has been found to be stable and has a Gaussian distribution during short-term intervals of 5 minutes with a relative standard deviation of 2.9 x 10^{-4}, which indicates that the electrometer voltage burden is stable for a calibrate – measure – calibrate procedure to be adopted. A switch is used to connect the electrometer to either the ion chamber or the electrical calibration system.

Using this system, it is possible to calibrate the electrometer with relative uncertainty better than 0.1 % over the range 10^{-12} A to 10^{-8} A, improving the measurement uncertainties and avoiding the risks associated with transporting the electrometer for calibration.

6. Summary and Outlook

We have described multiple promising avenues for the next generation of current measurement systems for ionization chambers. Work is already underway at some NMIs to incorporate the methods described above into existing ionization chamber systems and a joint CCRI-CCEM Task Group has been set up to address the topic (CCRI-CCEM, 2020). Beyond simply upgrading electronics, the fundamental change for many laboratories to defining a calibration factor for the

Bq based on a traceable SI ampere, no longer keeping a long-lived reference source “forever”, and measuring current with lower uncertainties creates new opportunities and challenges, requiring new quality assurance protocols. It is likely that reference sources will be lower activity, be changed more frequently, and be of shorter-lived nuclides, possibly having more impurities, than the historical ^{226}Ra sources used today (or until recently) at many NMIs and the BIPM. For these laboratories, low-uncertainty current measurements will become even more important for monitoring the long-term stability of IC measurement systems: dimensional and material stability, gas pressure, reference source decay behavior.

In the longer-term, one of the opportunities follows from recent developments in Monte Carlo simulations of ICs. Fully realizing a theoretical calibration in terms of absolute current will require better knowledge of the physical properties of the IC, such as gas pressure, and of physical constants inherent in the calculations, such as the mean energy absorbed per ion pair for the filling gas, which is typically known to only to about 0.6 % (de Vismes and Amiot, 2003).

In addition, there will be a need for greater understanding of lesser-studied dynamic effects such as diffusion, surface roughness, and other phenomena associated with ICs ageing over decades or centuries (Paepen, 2016). These same challenges will be of interest to quality assurance for current-based calibrations, whether based on traditional primary standards for activity, or theoretical calculations.

This discussion raises the issue of replacing the ionization chambers themselves, not just the measurement electronics - many chambers are at least 50 or 60 years old. It is likely that most practitioners will be reluctant to replace their chambers, due to the historical investment over many decades in determining calibration factors. An effort to design an ionization chamber that could be replicated stalled due, in part, to the difficulty of specifying and obtaining materials for

the walls of the chamber (Paepen, 2016). This latter difficulty might be related to the poorly-understood mechanism for charge recombination at the negative electrode (where the ions re-combine with electrons that have passed through the measurement circuit) (Schrader, 1997). If the electrode material is important, this suggests that overcoming this re-combination issue will involve research into materials physics and possibly electrochemistry.

While the authors have no solution to this problem, it is clear that a solution is necessary for this field to reach its potential - we encourage interested practitioners to start working on it!

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Disclaimer

Certain commercial equipment, instruments, or materials are identified in this paper to foster understanding. Such identification does not imply recommendation by the National Institute of Standards and Technology, nor does it imply that the materials or equipment identified are necessarily the best available for the purpose.

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FIGURES AND CAPTIONS

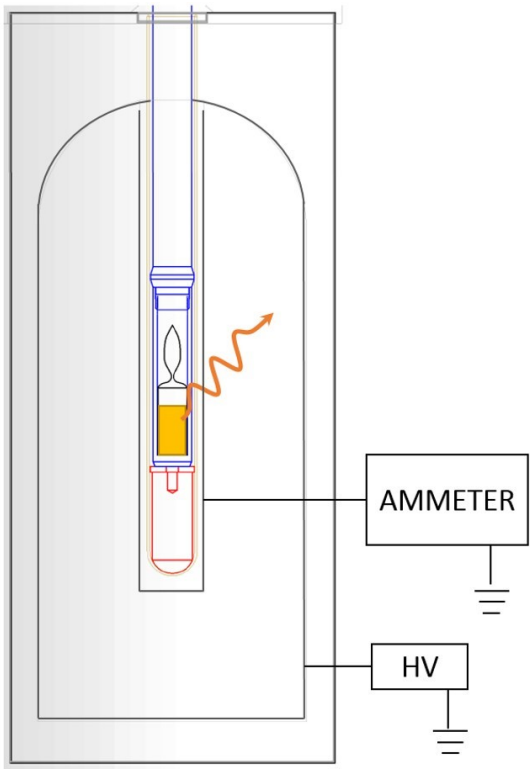

Figure 1 Schematic diagram of an ionization chamber, showing a radioactive solution (yellow shaded), which emits a γ-ray (orange arrow) that is absorbed in the pressurized gas region. The resulting current is measured with the ammeter.

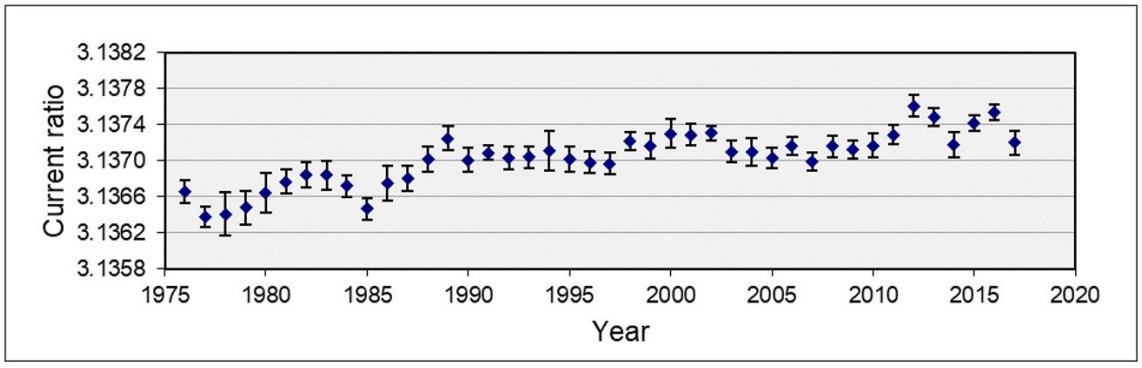

Figure 2 Ratio of the current produced by two ^{226}Ra sources as a function of time since 1976 at the BIPM, showing that a reproducibility of the order of 0.1 % can be achieved. However, a long-term trend in the ratio can be observed- the origin of this effect is unknown.

Figure 3 Schematic circuit diagrams of (a) an integrating electrometer, and (b) a feedback ammeter.

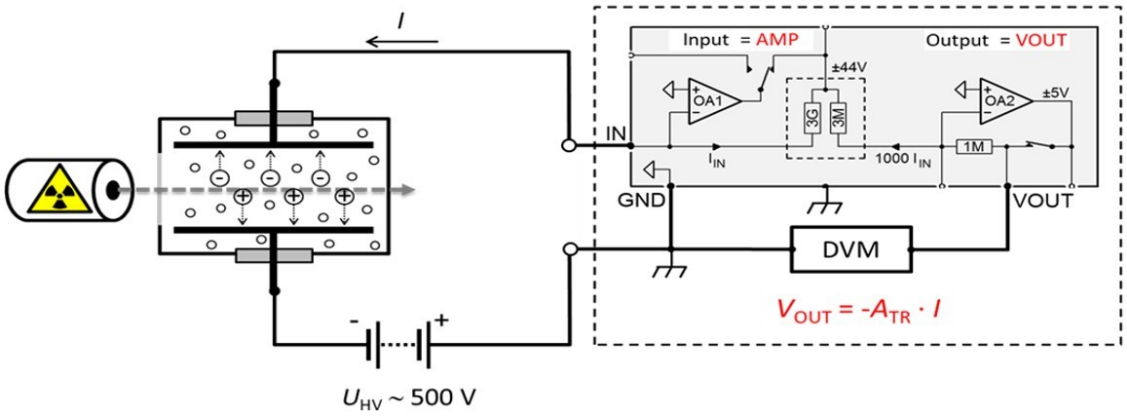

Figure 4 Example operation of a ULCA in an electrometer mode for current measurements on an IC (left), with A_{TR} = 1 GΩ (using the internal 1 MΩ resistor for current-voltage conversion) and for currents up to ± 5 nA. Note, the IC shown uses an external source, but the principle is the same for re-entrant ICs.

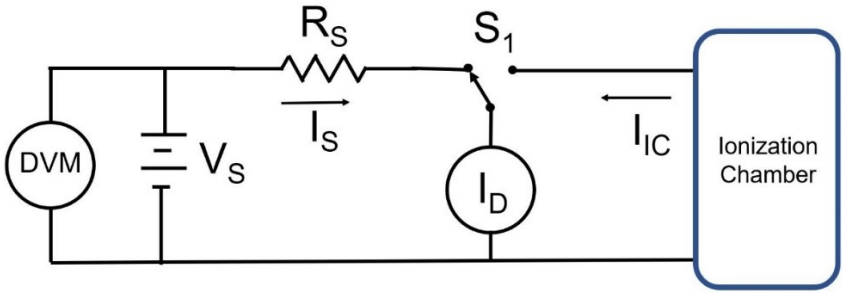

Figure 5 Circuit diagram for calibration of an electrometer used with an ionization chamber. Switch S1 connects the electrometer to the ionization chamber or the voltage source, DVM, and standard resistor. See text for definition of other symbols. A calibrate, measure, calibrate sequence is used to bracket ion chamber measurements with before and after calibrations of the electrometer.

TABLE AND CAPTION

Table 1 Some representative results of SEPs, showing maximum currents and relative uncertainties.

Author	**Journal**	**Year**	**Title**	**Material**	**Pumping Mode**	**I_{max} (pA)**	**Relative Uncertainty**
Stein	APL	2015	Validation of a quantized-current source with 0 .2 ppm uncertainty	GaAs	1-gate ratchet	87	2×10^{-7}
Yamahata	APL	2016	Gigahertz single-electron pumping in silicon with an accuracy better than 9.2 parts in 10^7	Si	1-gate ratchet	160	9×10^{-7}
Zhao	arxiv	2017	Thermal-error regime in high-accuracy gigahertz single-electron pumping	Si	1-gate ratchet	160	3×10^{-7}
Stein	Metro.	2017	Robustness of single-electron pumps at sub-ppm current accuracy level	GaAs	1-gate ratchet	96	2×10^{-7}
Keller	PRL	1996	Accuracy of Electron Counting Using a 7-Junction Electron Pump	Al/AlOx	pump	0.8	2×10^{-8}