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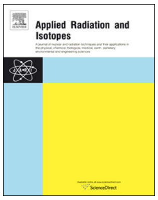

Linearity check of an ionisation chamber through $^{99\mathrm{m}}$Tc half-life measurements

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HIGHLIGHTS

- Measured $^{99\mathrm{m}}$Tc half-life of 6.00660 (18) h with ionisation chamber.
- Linearity of ionisation chamber confirmed over full dynamic range.
- Piecewise fit of decay curve gives same half-life value within 0.02%–0.04%.
- Integration of ionisation current over air capacitor assures better linearity.
- Direct current readout allows for wider dynamic range.

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ABSTRACT

The half-life of $^{99\mathrm{m}}$Tc was measured at the JRC using the ionisation chamber 'IC1' (type Centronic IG12). The result, $T_{1/2}(^{99\mathrm{m}}\mathrm{Tc}) = 6.00660$ (18) h, is in good agreement with literature data. One experiment was performed in IC1's default set-up with the ionisation current being integrated over an air capacitor and read out as a voltage increase over time. This ensured excellent linearity and precision throughout the dynamic range, but the maximum current was limited to 2 nA. In a second test, the current was directly read out with a Keithley 6517 A electrometer. Applying correction factors for the automatic range switching of the electrometer, an acceptable linearity was demonstrated over a range of 12 half-life periods starting at 20 nA. Range switching and autocorrelation of the current readout increase the systematic and random error propagation factors. Piecewise fitting of the decay curve over periods of 6 h yields the same $^{99\mathrm{m}}$Tc half-life value within 0.04% (0.0025 h) standard deviation over an activity range spanning at least 10 half-life periods (3 orders of magnitude).

1. Introduction

Re-entrant (or 4π) pressurized ionisation chambers (ICs) in combination with current-measuring devices are frequently used for secondary calibration measurements of radioactive sources in the fields of radionuclide metrology and nuclear medicine (Schrader, 1997, 2007). Owing to their favourable stability and linearity properties, ICs are well suited for the measurement of the half-life of gamma-emitting radionuclides. Nevertheless, for each instrument there are limitations to these properties depending on the specific configuration in which it is used. Good metrology requires that the validity of stability and linearity is checked to avoid erroneous conclusions from measurements. A striking example in that respect is the conjecture of 'neutrino-induced' beta decay on the basis of seasonal fluctuations in decay rate measurements, which could be refuted on the basis of a large-scale investigation comparing a multitude of instruments (Pommé et al., 2016, 2017a, 2017b, 2017c, 2017d, 2018a, 2018b).

Whereas the linearity range of nuclear counting devices is limited by pulse pileup and dead-time effects (Pommé et al., 2015), ICs can boast a wide dynamic range over which the ionisation current varies proportionally with the measured activity. Non-linearity effects may be caused by physical properties inside the IC – mainly incomplete charge collection due to local saturation and recombination of ion pairs – or by limitations in the electronic circuits producing the voltage/time or charge/time output (Schrader, 1997, 2007). Since half-life measurements are extremely sensitive to non-linearity effects (Siegert et al., 1998; Schrader, 2004; Pommé et al., 2008; Pommé, 2007, 2015, 2016), it is of importance to perform a linearity check over the dynamic range in which the instrument is used. At the JRC, half-life values for ^{65}Zn (Van Ammel et al., 2004), ^{54}Mn (Van Ammel et al., 2010), ^{124}Sb (Paepen et al., 2010), ^{109}Cd (Van Ammel et al., 2011), ^{177}Lu (Pommé et al., 2011), ^{134}Cs (Pommé et al., 2017b), and ^{22}Na (Pommé et al.,

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2017c) have been determined using 'IC1' (IG12, Centronic, UK), with currents ranging from a few pA up to 680 pA and a background current in the order of 0.045 pA.

In this work, a classical linearity check of IC1 is performed by following the decay curve of a short-lived $^{99\text{m}}$Tc (6 h) source over the dynamic range of the instrument. According to Schrader (2007), about five orders of magnitude of activity may be covered, and several hundreds of data points should be collected. An instrument of good linearity should show residuals to an exponential function to better than 0.1%. Such a procedure checks for the overall linearity of the measurement set-up, including the current readout. The $^{99\text{m}}$Tc half-life derived from this work will be compared to other measured values reported in the literature and recommended values from data evaluators.

2. Measurement conditions

2.1. $^{99\text{m}}$Tc source

The $^{99\text{m}}$Tc source material was obtained as sodium pertechnetate (TcO_4Na) in physiological saline solution, by elution from a ^{99}Mo-based technetium generator. About 10 GBq was provided to the JRC by the St.-Dimpna hospital of Geel, on Oct 15, 2012 for the 1st experiment and on Nov 5, 2012 for the 2nd. $^{99\text{m}}$Tc is commonly applied in hospitals for nuclear imaging in diagnostic procedures, making use of its 140 keV gamma ray emissions and conveniently short half-life. In the frame of the basic safety standards (EURATOM, 2014), there is a demand for proper calibration of all sources giving rise to medical exposure. The hospital calibrators are based on a similar design as the ICs used by metrology institutes. A recent intercomparison exercise among 15 Belgian hospitals showed that $^{99\text{m}}$Tc activities in vials and syringes are generally correctly reproduced within a margin of ± 5% (Saldarriaga Vargas et al., 2018).

2.2. IC1 with air capacitor

The IC1 is the principal ionisation chamber used for secondary calibrations of activity and half-life measurements at the JRC. It is a Centronic IG12 well-type chamber with 0.8-mm-thick steel wall, which is filled with argon gas at a pressure of 2 MPa. It is surrounded by a 50-mm-thick lead shield. The ionisation current is integrated over a custom-made air capacitor of 521.36 pF, which is placed in another lead shield to reduce discharge effects by radiation. The capacitor voltage is sampled by means of a Keithley 6517 A electrometer operating in voltage mode, triggered every 2 s by a stable crystal oscillator. The trigger period is measured with a calibrated frequency meter traceable to the SI unit second by comparison with a standard frequency generated by a DCF-77 receiver of which the oscillator is synchronised with the primary atomic clocks of PTB.

The raw data consist of voltage samples of the capacitor, taken every 2 s, in cycles from 0 to 9 V. When a voltage of nine volts is reached, the capacitor is discharged and after a waiting period of 30 s another charging cycle is started. For every two consecutive voltage samples, the voltage difference is calculated and these differences are checked for outliers (> 5 sigma), e.g. due to electronic spikes. If there is at least one outlier within a series of samples, 5 samples centred on the outlier are rejected from further processing. The incurred data loss is very small, whereas some unnecessary data scatter is avoided.

The duration of one cycle varies with the activity of the source. At currents exceeding 2.5 nA, the probability is high that the capacitor resets itself before the 2 s sampling period is completed. Below a current of 2 nA, each cycle contains at least one valid sample and this number grows inversely with the fading activity of the decaying source. At 1 nA there are 2 samples per cycle, at 800 pA there are 3, at 620 pA there are 4, and the number of samples goes up to about 51,000 in the last measurements. The experiment was pursued for 16 days, starting after a cooling period of 1 day on 16 Oct at a maximum current of 2074 pA.

Pre-treatment of the data consisted in taking averages of the voltage/time data and converting them to current via the capacitance (1 V/s corresponding to 521.36 pA). Considering the strong variation in cycle duration, a correction for decay during measurement was performed on the background-corrected current in each sample of a cycle, thus referring the mean activity to the starting point of the cycle. The pre-treatment equation used on the sample data is

$$I_{sample}\left(t,\, t_{cycle}\right) = \frac{\lambda_1 \Delta t}{1 - e^{-\lambda_1 \Delta t}}\left[C\frac{\Delta V}{\Delta t} - I_{bkg}\right] \tag{1}$$

in which t is the sampling time, t_{cycle} is the first sampling time of the cycle, $\Delta t = t – t_{\text{cycle}}$ is the decay time since the start of the cycle, λ_1 is the $^{99\text{m}}$Tc decay constant, ΔV is the voltage increase during Δt, and C is the capacitance relating current with change in voltage via $I = C\Delta V/\Delta t$, and $I_{\text{bkg}} = 0.0439$ pA is an adopted mean value for the background signal. The decay correction is valid in regions where the $^{99\text{m}}$Tc decay dominates, but somewhat distorts the decay curve at the end when the longer-lived ^{99}Mo and background signals take over. This is of lesser importance in the current context, but if needed, this effect can be reduced by taking shorter cycles or by adapting the data analysis procedure by including the inverse counting factors in the fit function of both nuclides and using the raw experimental data (see e.g. Pommé et al., 2011 and Collins et al., 2015).

2.3. IC1 with direct current readout

For a second experiment, the IC1 was decoupled from its standard set-up with air capacitor. Instead, the ionsation current was read out directly from a Keithley 6517 A electrometer in current mode. The electrometer was used in autorange mode, allowing it to change its settings according to the input current: at 200 nA, 20 nA, 2 nA, 200 pA, and 20 pA it switches to another feedback impedance. Data were sampled every 2 s and a time stamp was kept from the computer performing the data acquisition. The computer is continuously synchronised with the DCF-77 receiver by means of the network time protocol (NTP). Additional time information extracted from the electrometer was found to be inadequate for performing an unbiased half-life determination and therefore had to be excluded from use in the data analysis.

The experiment was started on Nov 5, 2012 and continued for 10 days. The ionisation current being sampled every 2 s created a set of 438106 (time, current) data vectors. The data set was not reduced in size and analysed as a whole, at the expense of long computation times.

3. IC1 with air capacitor

3.1. Fit of decay curve

In Fig. 1, the measured (background-subtracted) decay curve of $^{99\text{m}}$Tc is shown over the first 150 h (whereas the measurement was continued for an additional 150 h). The density of measurement points is high at the initial activity values and decreases gradually as the source decays, since more time is needed to charge the capacitor over the same voltage range. To the data set, a least-squares fit is performed of a model function consisting of two exponentials and a constant:

$$I(t) = A_1 e^{-\lambda_1 t} + A_2 e^{-\lambda_2 t} + B \tag{2}$$

in which λ_1 and λ_2 are the $^{99\text{m}}$Tc and ^{99}Mo decay constants, respectively, A_1 and A_2 are the corresponding amplitudes, and B is a constant which compensates for a possible residual bias in the background subtraction. The λ_2 parameter is kept fixed during the fit, using $T_{1/2}(^{99}\text{Mo}) = 2.7479\ (6)$ d (DDEP, 2004–2018). The contribution from ^{99}Mo activity is low, considering that the fitted amplitude ratio was $A_2/A_1 = 1.66\ 10^{-6}$ (noting that A_2/A_1 is a rough approximation of the