Purpose

The purpose of this procedure is to describe the measurement and reporting procedures for gamma-ray-emitting radionuclides using the NIST secondary standard ionization chambers (SSICs).

Scope

The NIST SSICs are "4π"–γ re-entrant pressurized ionization chambers, which are well-type instruments used for measuring the radioactivity (disintegrations per second) of gamma-ray-emitting radionuclides. Scheduled calibration services for submitted samples of gamma-ray emitters are offered as a complement to the Standard Reference Material (SRM) and Measurement Assurance programs. These calibration services are 43010C (half-lives longer than 15 days) and 43020C (half-lives shorter than 15 days). The nominal range of activity for the gamma-ray-emitting radionuclides is 0.4 MBq to 60 MBq and the nominal uncertainty range is 0.5 % to 2.3 % (expanded uncertainties, $k = 2$), both vary by radionuclide.

The SSICs use instrument-specific calibration factors (K-values) for each radionuclide previously established by direct methods of activity measurements (Hoppes 1984; NCRP 1985). Records of the direct measurements of activity used to determine K-values are kept in the K-value Record Book. For further information on specific methods refer to NIST SP 250-10. SSIC calibrations are checked with those of other national standardizing laboratories through international measurement comparisons, particularly those conducted by the Bureau International des Poids et Mesures (BIPM) with the Système de Référence (SIR), SIR Transfer Instrument (SIRTI), or Key Comparisons (https://www.bipm.org/en/radionuclide-metrology). Results of these comparisons are obtainable from the Key Comparisons Database (KCDB, https://www.bipm.org/kcdb/ comparison/quick-search) maintained by the BIPM.

The gamma-ray-emitting radionuclides which may be submitted to NIST for IC calibrations are those listed in the K-value Record Book as having established, primary standardizations

Definitions

K-value: The relative calibration factor, K_R, is given by R_R/R, the ratio of the measured responses of the ^{226}Ra reference source and a known one becquerel (1 Bq) of a stated radionuclide.

Equipment

Equipment manuals are maintained either as paper copies in Building 245, Room H223 or as electronic copies on the attached computers. Current instrument models and serial numbers are listed in the instrument-specific Lab Notebooks (ICA and AutoIC) located in H223. Current software and database versions are also listed in these notebooks and referenced by name and file date.

Hardware

• The NIST "4π"-γ (re-entrant) cylindrical ionization chamber “A” (ICA; described in NIST SP 250-10)
• The NIST Automated Ionization Chamber (AutoIC; also referred to as ionization chamber “B”, ICB) [Described in Fitzgerald (2010)]
• Multi-range electrometer (for each SSIC)

• Sample holder(s)
• Radium-226 reference sources
• A network connected desktop computer

Equipment Quality Control

Multi-range electrometer: Individual measurements of submitted sources are evaluated as ratios to the appropriate reference source, therefore normally fluctuating environmental conditions do not impact the measurement result. Multi-range electrometers may be subject to non-linearities between ranges. Upon installation correction factors are measured based on a short-lived decaying source or a variable current source and, if appropriate, incorporated in the data collection program. The linearity of the electrometers is checked biennially, as scheduling allows, not to exceed 2.5 years, using short-lived decaying sources. The result of a check for consistency, including any updates to correction factors, is recorded in the IC lab notebook.

Radium-226-reference sources: K-values are sometimes determined for only one RRS. K-values for other RRSs are calculated using RRS ratios. RRS ratios are checked at least every five years. The result of the measurements and comparison with historical data is recorded in the IC Lab Notebook. Failure of an individual RRS should be investigated if there is a change in ratio greater than three times the standard uncertainty on the ratio measurement. RRSs are checked for external contamination, indicating possible failure, prior to each measurement campaign.

Software

Data acquisition software
ICi_[x].vi: ICi is a LabVIEW program running on the ICA desktop computer. The current version of the software is linked by a desktop shortcut. It is used to collect data from the ICA with input from the user. Data are stored offline for further processing. Data are backed up in accordance with the Data Management Plan for ICs.

Robot.exe: Robot is a Visual Basic program running on the AutoIC desktop computer. It calls the current version of a LabVIEW program that collects data from the AutoIC with input from the user. Data are stored offline for further processing.

Data analysis software

Ionol4.exe: Ionol is a FORTRAN program running on a desktop computer. It is used to process the data file created by the acquisition programs using further input from the user and information from ICDAT4.TXT.

ICDAT4.TXT: ICDAT4.TXT is a data file containing the most recent K-values listed in the K-value Record Book for both the ICA and AutoIC. Note, the ICA K-values listed in this file have been corrected for the height effect (Fitzgerald 2012), as documented in the K-value Record book. For each radionuclide, the database also includes the half-life to be used for any decay corrections by the analysis software.

Validation

The validation of equipment is performed upon installation and subsequent to any change in equipment, and subsequent to any significant environmental event.

Validation of processing software is performed by: manual calculation of an experimental result and comparison of this value to that obtained from the program or processing of a test data set yielding a known activity result. This is performed upon the initial version and subsequent to any changes in the program.

Results of validations are recorded in the IC Lab Notebooks.

Safety

Radiation safety

Radiation safety training and assessment services are provided by the NIST Radiation Safety Division (RSD). The room used for ionization chamber calibrations is designated as a Radiation Area. A countertop lead cave is used to house samples during measurement and to store the ^{226}Ra reference sources. In the case of high-activity sources, a lead-lined “L” shield might be used while changing samples.

Ampoules to be measured must be checked and be found to be free of any external radioactive contamination or defects in the flame-seal before being loaded into the ionization chamber sample holder.

Procedures

Source Container and Contents

The solution to be assayed in the SSICs should have as nearly as possible the same composition as the standards used to calibrate the chamber (described in NIST SP 250-10). The gamma-ray-

emitting radionuclide solution should be (5.0 ± 0.2) mL in a standard, NIST-type, 5 mL ampoule. The specifications of these ampoules are as follows:

body diameter	16.5 ± 0.5 mm
wall thickness	0.60 ± 0.4 mm
barium content	less than 2.5 percent
lead oxide content	less than 0.02 percent
other heavy elements	trace quantities

Description of the Actual Measurement Process

The stability of these chambers is monitored by measuring response to a reference source of ^{226}Ra in equilibrium with its daughters whenever calibrations are made. The activity of the sample is taken to be proportional to the ratio of the chamber response (current) for the sample to that for the reference source. The relative calibration factor, K_R, is given by R_R/R, the ratio of the measured responses of the ^{226}Ra reference source and a known one becquerel of a stated radionuclide.

The activity, A, of the radionuclide in a sample to be measured, S, is given by $K_R R_S$, where R_S is the response (current) for the sample relative to that for the radium source. Background currents, measured just before and after counting the sample and radium, must be subtracted from the response for each. The K_R diminishes from the value at the time of the original calibration with the 1600-year half-life of the ^{226}Ra, and a decay correction for this must be made.

Preliminary

• Customer contact: give specifications for chemical composition and activity limits, source must either be provided as 5 mL solution in a NIST-type 5 mL flame-sealed ampoule, or transferred to one by NIST.

• NIST paperwork and acceptance procedure: submit completed NIST 364 (or NIST 365), Radioactive Material Request, for approval before arrival of materials. Specifically notify the Chief of RSD or designate of the arrival and departure of all isotopes of plutonium and uranium. Complete any other necessary forms (NIST 64, Test Record; NIST 77, Calibration and Test Fee Computing Form; NIST 796A, Shipper's Declaration for Radioactive Materials, etc.).

• E-Commerce: Note dates of material received (and returned) in the E-commerce system.

• Schedule time to use the IC's by blocking and initialing the required amount of time on the calendar in 245/H223. Possible conflicts should be resolved in person with other users. Generally shorter half-life sources take precedence.

• Deliver source, or a related sample, to the appropriate person to be measured on a calibrated germanium spectrometry system according to instructions for an impurity measurement. The results and source are delivered to the analyst at the completion of this measurement.

Receiving Instructions for Radioactive Solution Samples

All radioactive sources arriving at NIST are delivered to RSD where they are examined to determine the radiation level and possible contamination of containers and packaging.

After RSD releases the source to the analyst, the analyst in turn should verify that the correct radionuclide has been shipped in the activity range which has been previously discussed with the customer. Quality control procedures that should be implemented by the analyst are as follows:

1. Re-examine the source for pin holes of any type and check the ampoule seal.

2. Inspect the label attached to the primary container. This helps to ensure that it is the expected radionuclide and the activity amount is correct.

3. Read technical data sheets and package inserts accompanying the radionuclide package carefully for special instructions on stability, quality inspection, and chemistry of the product. This information should conform to the calibration guidelines specifying the physical and chemical properties of the submitted sample solution.

Providing that no defects are observed and clean swipes have been documented by RSD or by the analyst, the sample may be transported for measurement in the SSICs. In the case of above criteria not being met, contact customer.

Step-by-Step Operating Instructions for the SSICs

Complete an IC Log Book entry.

<u>Starting the SSIC system</u>

The SSIC system to be used is started from its connected computer using the following procedure:

1. Check schedule and observe system to verify it is not currently in use.

2. Verify computer system time is set to the correct Eastern Standard Time (EST). If necessary, correct the time using NIST network time (check nist.time.gov and be sure to convert to EST if checked during daylight saving time).

3. For ICA, click on desktop icon ICi_[x].vi. For AutoIC, click on desktop icon Robot.exe. and click “OK” when prompted.

Background Measurements

Background measurements are made initially and should be examined before continuing. If the background is inconsistent, or abnormally different, investigate the cause. High background may be due to an unshielded source in the vicinity. It is possible to make measurements under high background conditions by measuring the background more often. Typical backgrounds can be viewed using the program Control Chart Viewer.vi on the respective attached computer.

Loading Sources

Complete procedures for safely handling radioactive sources and placing them into the source holders, including PPE and tools, are provided in the appropriate SOPs.

When placing the solution sample in the chamber, observe the ampoule to make certain that no liquid is in the neck of the ampoule.

ICA
Gently place the ^{226}Ra reference source or ampoule into the rigid, tight-fitting Lucite source holder ("holder 2"). With the bottom part of the holder seated into the top part, align the markings on the RRS with the alignment mark on the source holder. Gently lower the source holder into the chamber using the string; do not drop. Align the markings on the source holder with those on the instrument shielding. (Note – The source holder is an integral part of the IC system. Care must be taken to place the holder shaft in a position where it will not roll and fall. If a cart is used, it must have walls or a tray to restrict movement of the holder.)

AutoIC
Gently place the ^{226}Ra reference source and/or ampoule(s) into rigid, tight-fitting source holders, screw the top and bottom pieces together taking care not to cross-thread or overtighten, and place them in appropriate positions on the AutoIC source rack.

Measuring the RRS and Sample(s)

Alternate measurements of RRS and samples. Each measurement duration is typically set for at least 60 seconds, with longer times used for weaker sources. At least four sets of 10 measurements should be made on the sample, with bracketing measurements made on the radium reference source in each cycle.

For ICA, each measurement requires completing the appropriate fields (highlighted in blue) in the software for each source before each measurement. For AutoIC, the parameters for all of the measurements to be performed are entered by setting up a sample database in the Robot.exe software.

To commence ICA measurements, click "Start Run". To commence AutoIC measurements, click "Run samples using database".

After all measurements have been completed, it is recommended that background measurements be repeated.

Data Collection

The data collection is under control of data acquisition software. All data are collected and stored to a disk for off-line analysis. Data are backed up in accordance with the Data Management Plan Pibida gamma spec.

Measurement of impurities

The following gives a summary of the procedures documented in the *Procedure for gamma-ray spectrometry measurements for activity calibration and impurity measurements using High Purity Germanium (HPGe) detectors.*

Impurity-check measurements of radioactive sources to be calibrated by SSICs are performed using the Radiation Physics Division (RPD) Gamma-Ray Spectrometry Systems located in Building 245, Rooms B119 and H119. This system consists of six High Purity Germanium (HPGe) detectors and one Lithium-drifted Silicon detector. These detectors are calibrated using NIST Standard Reference Materials (SRMs), which are, in turn, validated through measurement comparisons with other national metrology institutions and the BIPM through the periodic submission of measured sources to the SIR for gamma-ray emitting sources. The calibration of each of the seven detectors is done through the development of efficiency curves for different source geometries and various source-to-detector distances. These efficiency curves are verified monthly or more frequently if needed (such as when there has been a change in the instrument configuration), using previously calibrated ^{57}Co and ^{60}Co point sources to determine the efficiency value, shape (FWHM) and energy calibration reproducibility. Efficiency curves for point sources (solids), 5 mL ampoules (liquids) and 33 cm^3 spheres (gas) for gamma-ray emitting sources for the energy range 14 keV < E < 3.6 MeV are available for impurity and activity calibration measurements.

Gamma-ray spectra are acquired using GammaVision™ (version 8.00.03 as of this writing), a commercially available software developed and marketed by Ametek/EG&G ORTEC or using Genie 2000 from Canberra (Version 3.4.1) depending on the detector. Data analysis is performed using Genie 2000 from Canberra (Version 3.4.1). The choice of data analysis software was based on in-house comparisons of the results obtained by several (4) different software packages. In addition, the data analysis software used by customers was taken into account. (Such commercial products are identified in order to specify these procedures accurately; the RPD does not recommend or endorse any commercial product, nor intend to imply that these products are necessarily the best available for the purposes of these procedures.)

The combined standard uncertainty (u_c, corresponding to k = 1) on the activity values for the impurities and/or source measurement is given by

$$u_c = \sqrt{u_{\text{eff}}^2 + u_{\text{measure}}^2}$$

where u_{eff} is the uncertainty of the efficiency for the measured gamma-ray energies for the impurity and/or source, and $u_{\text{measure}}$is the uncertainty calculated based on the gamma-ray lines used to determine the activity of the measured impurity and/or source. As impurity levels are generally much lower than the nuclide being calibrated, the uncertainty on the impurity measurement is often on the order of 5 % to 20 %, for very small impurity levels the uncertainty maybe higher.

Processing the IC Data

The data processing is done using Ionol4.exe. The program averages the data for each sample and corrects them for background and radioactive decay (using the established half-life). The sample activity is calculated using the K-value for that radium reference source most closely producing a current approximating that of the sample. The results are printed out.

Correction for impurities is handled by the processing program. Results of impurities are entered as an activity ratio relative to the main radionuclide and a measurement time.

In specific cases, documented in the K-Value Record Book, the K-value may be dependent on solution composition, particularly the density of the solution. This is normally seen in low-energy gamma-emitters. If the solution composition of the measured sample is different from the composition of the sample used to determine the K-value, a correction to the K-value must be made using the equation listed in the K-value Record Book.

Acceptance Criteria

The data are examined to see if the results show the expected activity, and the standard deviation of repeated measurement on the source is checked to see if it is in the range expected for the radionuclide (normally less than 0.1 percent at 1 sigma). If there is a discrepancy in activity, the source may be measured the other SSIC. Alternatively, the source may be measured in a commercial re-entrant ionization chamber, or “dose calibrator,” as a confirmatory measurement. This measurement is performed by placing the sample in the dose calibrator source holder and entering the correct instrument setting for the radionuclide in question. Settings are found on a list attached to the instrument or in the instrument manual. The analyst must be familiar with the use of the dose calibrator and the accuracy of the settings. If the standard deviation is abnormally high, the data is examined for possible trends. The customer is contacted if a discrepancy cannot be resolved.

Calibration Report

At the end of a calibration exercise, the customer will be given a Report of Calibration stating primarily:

a) the principal radionuclide

b) reference time and date

c) SSIC used and method of calibration (traceability to primary standard)

d) certified values for activity

e) decay-scheme assumptions

f) assessment of radionuclidic purity

g) overall uncertainty determinations for calibration

Reported values are checked against the data on the IC printout and this check is noted by initialing the printout. Drafts of reports are stamped as such and identified by draft number. Results of draft proofreading are noted and initialed on the draft.

Signatures are obtained at the appropriate level and, if paper, the original report is embossed with a Department of Commerce seal. A copy marked file copy and two marked copies are maintained with the calibration printouts, stored in folders identified by company, radionuclide, date, and service number. If requested, the original paper report is sent to the customer. Electronically signed reports are stored in the calibration record of the E-commerce system.

Customer Sources

When the calibration service is completed, the technical users are notified, and the calibrated sample returned, if desired by customer. Short-lived radionuclide samples are usually measured by the customer only before transmittal to NIST, for the activity would be too low for measurement after the NIST calibration.

Uncertainty Analysis

The bases for the determination of uncertainties are the ISO *Guide to the Expression of Uncertainty in Measurement* and the *Guidelines for Evaluating and Expressing the Uncertainty of NIST Measurement Results*.

The purpose of this section is to explain the derivation of the various components of uncertainty. Measurements performed with a SSIC must be done under completely fixed conditions. This requires a stable background, no saturation losses due to incomplete charge collection because of

recombination at high rates, and corrections for all impurities. Ampoules must be of the NIST standard 5 mL type, the volume in ampoules must be 5.0 ± 0.2 mL, and the ^{226}Ra reference source must be corrected for decay.

Uncertainty in activity measurements may be increased if the contribution of the background is variable, large, or both. The uncertainty quoted is derived from measurement of the variations. The operator of the chamber should make frequent measurements of the background and look for the source of any unusual variations observed.

• K-value (Type A and Type B) – uncertainty in the determination of the K-value, including the measurement of activity by primary methods and measurements to transfer that calibration to the ionization chamber (specific uncertainties for individual K-values are listed in the K-value Record Book)

• RRS ratio (Type A) - uncertainty in the ratio of radium reference sources, when the radium source used in the measurement is different from the radium source used for the determination of the K-value

• Half life (Type B) – uncertainty in the half-life value(s) used for decay corrections

• *N* measurements on this sample (Type A) – standard deviation determined from *N* measurements of activity ratios

• Impurities in this sample (Type A and Type B) - Radionuclidic impurities may or may not have a significant effect, depending on the radiations of the principal and impurity radionuclide and the amount of impurity present. If their half-lives are relatively short, measurements at different times may be used to check consistency. High purity germanium detector systems are used to identify and quantify the significant radionuclidic impurities that may be present. The uncertainty in these measurements is propagated through the calculated effect on the IC results.

• Gravimetric (Type B) – typical uncertainties from gravimetric transfers of solution, if necessary

• Density (Type B) – uncertainty from correction for differences between density of submitted source solution and the solution used to determine the K-value, if necessary

Records

Calibration printouts and report copies are stored in folders identified by company, radionuclide, date, and service number.

If the source is returned, the date that the source is shipped back to the customer should be entered into the folder.

Folders of calibration records are maintained in 456/C117 for at least five years.

Documentation

K-Value Record Book – maintained in 456/C115

Ionization Chamber Laboratory Notebooks – maintained in 245/H223

Ionization Chamber Log Book (tabs for ICA and AutoICs) – maintained in 245/H223

SSIC software manuals – maintained in 245/H223

Standard Operating Procedures for SSICs – 846.04-0035 and 846.04-0037 posted in the 245/H223

Procedure for gamma-ray spectrometry measurements for activity calibration and impurity measurements using High Purity Germanium (HPGe) detectors – maintained in 245/B119 and 245/H119.

References

Hoppes, D. D., Basic Radionuclide Measurements at the United States National Bureau of Standards, Environmental Int. 10, 99-107 (1984).

NCRP Report 58, A Handbook of Radioactivity Measurements Procedures,

National Council on Radiation Protection and Measurement, Bethesda, Maryland,

(1985).

Taylor, B.N., and Kuyatt, C.E. “Guidelines for Evaluating and Expressing the

Uncertainty of NIST Measurement Results,” NIST Technical Note 1297 (1994).

Calhoun, J.M., Radioactivity Calibrations with the NIST “4π” Gamma Ionization Chamber, NIST Special Publication 250-10, 1987

Fitzgerald, R. An automated ionization chamber for secondary radioactivity standards, App. Radiat. Isotop. 68, 1507 (2010).

Fitzgerald, R. NIST Ionization Chamber “A” Sample-Height Corrections, J Res Natl Inst Stand Technol Vol 117 (2012).

Filing and Retention

Calibration printouts are stored in folders identified by company, radionuclide, date, and service number.

If the source is returned, the date that the source is shipped back to the customer should be entered into the folder.

For customer calibration, prepare calibration report and obtain required signatures. Make a copy for customer file and send original. Reports created and signed digitally are downloaded to the calibration record in the E-commerce system.

The RPD Quality Manager shall maintain the original and all past versions of this RPD Procedure.

Appendix A. Sample Calibration Report

National Institute of Standards & Technology

REPORT OF CALIBRATION

for

COMPANY, INCORPORATED
TOWNESVILLE, STATE

Radionuclide	Linolium-100
Source identification	1234-abc
Source description	Liquid in a NIST borosilicate-glass flame-sealed 5-mL ampoule (1)*
Solution composition	$LoCl_2$ in 0.1 mol·L^{-1} HCl (2)
Activity	1.235 x 10^5 Bq
Expanded uncertainty (k=2)	1.5 percent (3)
Reference time	1200 EST 30 February 2018
Photon-emitting impurities (Activity ratios at reference time)	$^{101}No/^{100}Lo$: (1.2 ± 0.3) x 10^{-5} (4)
Half life	(123.45 ± 0.06) days (5)
Measuring instrument	NIST "4π"-γ ionization chamber A calibrated by apple π(e+x)-γ coincidence counting (6)

Measurements Performed by

Jeffrey T. Cessna, Physicist

For the Director,
National Institute of Standards and Technology by

Brian E. Zimmerman, Acting Leader
Radioactivity Group
Physical Measurement Laboratory

Michael G. Mitch, Acting Chief
Radiation Physics Division
Physical Measurement Laboratory

Gaithersburg, MD 20899
Report Issued: 1 April 2018
Service ID No.: 43010C
NIST Folder No.: xxxxx-xx

*Notes on back

NOTES

(1) The solution was prepared by Company, Incorporated (CI), Townesville, State. CI reported the mass to be 4.9938 grams.

(2) Description provided by CI.

(3) The uncertainty analysis methodology and nomenclature used for the reported uncertainties are based on uniform NIST guidelines and are compatible with those adopted by the principal international metrology standardization bodies [cf., B.N. Taylor and C.E. Kuyatt, *NIST Technical Note 1297* (1994)].

The combined standard uncertainty, u_c = 0.75 percent, is the quadratic combination of the standard deviations (or standard deviations of the mean where appropriate), or approximations thereof, for the following component uncertainties:

a) 40 ionization-chamber measurements on this sample	0.02 percent
b) K - value	0.72 percent
c) photon-emitting impurities in this sample	0.02 percent
d) radium reference sources ratio	0.08 percent
e) radium reference source positioning	0.10 percent

The expanded uncertainty, U = 1.5 percent, is obtained by multiplying u_c by a coverage factor of k = 2 and is assumed to provide an uncertainty interval of approximately 95 percent confidence.

(4) Limits of detection for impurity gamma rays are:

1.2×10^5 $\gamma \cdot s^{-1}$ between 30 keV and 110 keV,
3.8×10^4 $\gamma \cdot s^{-1}$ percent between 130 keV and 350 keV, and
4.6×10^3 $\gamma \cdot s^{-1}$ percent between 400 keV and 1800 keV,

provided that impurity photons are separated in energy by five keV or more from those emitted in the decay of linolium-100. Limits are as of impurity measurement time, 32 January 2015.

(5) Evaluated Nuclear Structure Data File (ENSDF), February 2015.

(6) Ionization chamber calibration is performed using a primary measurement method, thereby establishing traceability to NIST standards for measurement of activity of this radionuclide (Calhoun, J.M., Radioactivity Calibrations With the NBS 4$\pi\gamma$ Ionization Chamber, and Other NBS Radioactivity Calibration Capabilities (NBS SP 250-10), Gaithersburg, MD: National Bureau of Standards, October 1987).

For further information, please contact Jeffrey T. Cessna at (301) 975-5539.

NIST Folder No.: xxxxx-xx
Source Identification: 1234-abc