Source self-attenuation in ionization chamber measurements of ^{57}Co solutions

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Abstract

Source self-attenuation for solutions of ^{57}Co of varying density and carrier concentration was measured in nine re-entrant ionization chambers maintained at NIST. The magnitude of the attenuation must be investigated to determine whether a correction is necessary in the determination of the activity of a source that differs in composition from the source used to calibrate the ionization chamber. At our institute, corrections are currently made in the measurement of ^{144}Ce , ^{109}Cd , ^{67}Ga , ^{195}Au , ^{166}Ho , ^{177}Lu , and ^{153}Sm . This work presents the methods used as recently applied to ^{57}Co . A range of corrections up to 1% were calculated for dilute to concentrated HCl at routinely used carrier concentrations.

Key words: ionization chamber, self-attenuation, ^{57}Co solution

1. Introduction

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Source self-attenuation is a known problem for low-energy gamma-ray emitters when being measured in ionization chambers. Normally the recommendation is to keep carrier and acid concentrations low, resulting in solutions of densities approaching $1 \text{ g}\cdot\text{mL}^{-1}$. Identical source geometry and solution compositions are to be used in the calibration of the ionization chamber as in the later measurements of the same radionuclide (Schrader 1997). This is not always practical when offering a calibration service or when investigating drug products. Either the ionization chamber must be recalibrated with an identical geometry or solution composition or a correction factor must be determined. NIST has previously investigated the effect of varying geometry for ^{57}Co calibrations (Calhoun et al. 1987). The magnitude of the attenuation must be investigated to determine whether a correction is necessary in the determination of the activity of a ^{57}Co source that differs in composition from the source used to calibrate the ionization chamber. At our institute corrections are currently made in the measurement of ^{144}Ce , ^{109}Cd , ^{67}Ga , ^{195}Au , ^{166}Ho , ^{177}Lu , and ^{153}Sm . This work presents the methods used as recently applied to ^{57}Co .

2. Materials and Methods

Sources were gravimetrically prepared by mixing various proportions of distilled H_2O , concentrated HCl (Baseline, Seastar Chemicals, Sidney, Canada)[†], a concentrated cobalt

[†] 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.

carrier solution, and approximately 0.5 mL of a ^{57}Co standard solution, #2355. The carrier and standard solutions were gravimetrically prepared at NIST. The activity concentration of a gravimetric dilution from the standard was determined using the live-timed anticoincidence counting method (Bobin, 2007). A series of 10 ampoules were prepared representing a range of HCl molarity from $0.01 \text{ mol}\cdot\text{L}^{-1}$ to $9.3 \text{ mol}\cdot\text{L}^{-1}$, corresponding to a density range from $0.998 \text{ g}\cdot\text{mL}^{-1}$ to $1.144 \text{ g}\cdot\text{mL}^{-1}$. The carrier concentration ranged from $14 \text{ }\mu\text{g} [\text{Co}]$ to $2.5 \text{ mg} [\text{Co}]$ per gram of solution. Individual characteristics are given in Table 1. All sources contained approximately 5 mL solution in a standard NIST 5 mL flame sealed ampoule. The activities ranged from 8 MBq to 13 MBq ^{57}Co at time of measurement. Additionally, the sources contained ^{56}Co and ^{58}Co as impurities. The ratio of the impurities to the ^{57}Co were $(4.10 \pm 0.53) \times 10^{-4}$ and $(1.90 \pm 0.07) \times 10^{-4}$, respectively, at the reference time ($k=1$). The reference time was within a day of the ionization chamber measurement times.

Measurements were made using the following ionization chambers: the NIST secondary standard $4\pi\gamma$ ionization chamber “A” (ICA) (Calhoun 1987); the Vinten 671 (VIC) (Woods et al. 1983); AtomLab models 400 (AL-400) and 500 (AL-500); and Capintec models CRC-15, CRC-15R, CRC-35R, CRC-25PET, and CRC-1.8atm (The 1.8-atm chamber from Capintec is the basis of their CRC-PC dose calibrator.). For the ICA, measurements were made relative to the response on a radium-226 reference source, #50. The current was measured in the VIC using an electrometer under computer control. The CRC-15R, CRC-25PET, CRC-35R, CRC-1.8atm, and AL-400 chambers were read by

computer. In both cases control was by NIST-written LabVIEW programs. The CRC-12 and AL-500 readings were taken by eye from the instrument display.

Uncertainties in the readings are standard deviations of 40, 400, or 10 repeated readings, respectively for the ICA; VIC; and CRC-15R, CRC-25PET, CRC-35R, CRC-1.8atm, and AL-400. For the CRC-12 and AL-500, the uncertainty is assigned based on an observation of the range of values during the visual average of the display to assign a value.

Instrument readings were corrected for background and decay corrected approximately 8 days to a common reference time using a ^{57}Co half-life of 271.80(5) days. Impurities were decay corrected using half-lives of 77.240(26) days and 70.85(3) days for ^{56}Co and ^{58}Co , respectively. All half-lives are from DDEP (Bé et al, 2004).

3. Results and discussion

All reported uncertainties are standard uncertainties unless otherwise indicated. Reported results are only considered valid over the range of densities and carrier concentrations studied, as extrapolation would introduce additional uncertainties, which were not considered.

3.1 Secondary standard ionization chambers

The instrument response for each source was divided by the mass of solution #2355 it contained in order to make comparisons on a relative basis. A least squares fit was performed to the following equation:

$$R = a + b * \rho + c * [Co] \quad (1)$$

where R is the ionization chamber response, ρ is the solution density in $\text{g}\cdot\text{mL}^{-1}$, and $[Co]$ is the carrier concentration in terms of $\text{mg}\cdot\text{g}^{-1}$ of Co. The coefficients of the fit are a , b , and c . Dividing the coefficients by the result using the characteristics of the standard solution gives a new equation with a_c , b_c , and c_c , the inverse of which can be used as a correction factor for composition, C_C :

$$C_C = \frac{1}{a_c + b_c * \rho_{\text{sol}} + c_c * [Co]_{\text{sol}}} \quad (2)$$

where ρ_{sol} and $[Co]_{\text{sol}}$ are the characteristics of the solution to be measured in units as above.

Figure 1 shows the response in ICA, as a ratio to the ^{226}Ra reference source (RRS), with respect to the solution density and carrier concentration. A clear relationship is seen between increased solution density and decreased response in the ionization chambers. A smaller decrease can be identified with respect to an increase in carrier concentration.

The coefficients a_c , b_c , and c_c , to calculate C_C for ICA are 1.0577, -0.0567, and -0.0001, respectively. For the sources in this experiment the magnitude of the resulting correction ranges from -0.12% to 0.71%. For a realistic solution of 1 mg $[Co]$ per gram $4 \text{ mol}\cdot\text{L}^{-1}$ HCl, approximate density $1.066 \text{ g}\cdot\text{mL}^{-1}$, the magnitude of the correction would be 0.28%.

This correction is more than half the magnitude of the combined uncertainty on the ICA k -value for ^{57}Co of 0.41%.

The uncertainty on this correction is based on components for the average uncertainty in: repeated measurement of a single sample, 0.02%; fit of the data, 0.01%; mass of the contained activity, 0.05%; and components for the uncertainties in the ρ and $[\text{Co}]$ in the standard and the measured source. The uncertainty on the density of the standard of 0.16% results in a 0.01% uncertainty on the correction. The sensitivity for uncertainty in carrier concentration is 0.0001, making the component negligible. In the example above if the ρ_{sol} and $[\text{Co}]_{\text{sol}}$ are known to 0.25%, the resulting combined uncertainty would be 0.06%. If the ρ_{sol} and $[\text{Co}]_{\text{sol}}$ are only known to 5%, the resulting combined uncertainty would be 0.3%, with the uncertainty on the density of the unknown being the dominant component. Therefore, knowledge of the uncertainty on the density is important. If the density, or the uncertainty on the density, is poorly known, the method may still be used to set a limit on the possible introduced bias in measured activity.

In the results for the VIC, the effect of $[\text{Co}]$ is even smaller, giving a c_c of 0.00003, and can be ignored. The values for a_c and b_c are 1.0645 and -0.0628, respectively. These values indicate a slightly larger effect with respect to density. The corrections range over 1.2%. Using the example measurement above yields a correction of 0.34% with a combined uncertainty of 0.10% on the correction. Again, a comparison can be made to the uncertainty in the k_{VIC} of 0.42%.

3.2 Radionuclide “dose” calibrators

The coefficients of the equation for C_C and with the results of the example case and the associated uncertainty are given in Table 2. The secondary standard ionization chambers are given for comparison.

When evaluating the utility of the correction for dose calibrators one must consider the uncertainty in the dial setting for ^{57}Co . Manufacturers give a nominal uncertainty of 2% to 3% on their dial settings. This is obviously far larger than the correction to be made. In practice, dose calibrators can be individually calibrated with a smaller uncertainty, on the order of 1% of activity. In this case a correction can be made. In either case, an uncertainty component on the activity from the instrument can be estimated for the effect from solution density.

One must also consider how well the data fit equation 1. This can be achieved by examining the residuals of the fit. In some cases the data are acceptable. The coefficients a_c and b_c , to calculate C_C for CRC-15R are 1.0571 and -0.0566, respectively. The resulting corrections range over 0.8%. The example case yields a correction of 0.34% with a combined uncertainty of 0.19% on the correction.

The corrections using the example case are as large as 0.53%, for the AL-400. In some cases the uncertainties on the underlying data did not allow a good fit. For the CRC-12, CRC-35R, CRC-25PET, and the AL-100 the uncertainty is on the order of or larger than

the correction. In the example case this uncertainty can be used to set a limit on the effect of the density on the ionization chamber response.

4. Conclusions

The effect of source self-attenuation on ionization chamber response was investigated for solutions of ^{57}Co of varying density and carrier concentration. The implementation of a correction factor is warranted in those chambers used as secondary standards. For the NIST ICA and VIC chambers, an equation to derive a correction factor as a function of solution density was given. The resulting corrections are generally $< 1\%$ and have associated uncertainties $< 0.1\%$ on the correction. In the case of radionuclide activity calibrators the uncertainty in determining a correction factor can be of the same or larger magnitude than the correction, particularly if the density of the measured solution is not well known. From an examination of the maximum variation of the data over the range studied, a limit can be placed on the expected variation due to solution density of between -1.03% and $+0.57\%$, depending on the chamber. The results are valid for a density range a density range from $0.998\text{ g}\cdot\text{mL}^{-1}$ to $1.144\text{ g}\cdot\text{mL}^{-1}$.

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Figure Captions

Figure 1

The response in ICA, as a ratio to the RRS, with respect to the solution density and carrier concentration. The projected points are the experimental data. The gridded surface represents a fit of equation 1 to the data.

Figure 1

Table 1

Individual source characteristics for the series of ampoules used in this experiment.

Densities are given at 20 °C.

Source	Total Mass (g)	Total Volume (mL)	Density (g·mL⁻¹)	[Co] (mg·g⁻¹)	[HCl] (mol·L⁻¹)
#2355-1	5.19435	5.15173	1.0083	2.462	0.015
#2355-2	5.48528	4.90166	1.1191	1.552	7.200
#2355-3	5.02972	5.03763	0.9984	0.0163	0.011
#2355-4	5.38188	4.99989	1.0764	1.181	4.621
#2355-5	4.97228	4.93023	1.0085	2.541	0.011
#2355-6	5.58666	5.04382	1.1076	2.282	6.279
#2355-7	4.99637	5.00418	0.9984	0.0169	0.011
#2355-8	5.69406	4.97760	1.1439	0.0153	9.202
#2355-9	5.65255	4.93678	1.1450	0.0143	9.268
#2355-10	5.04918	4.97292	1.0150	0.0934	1.060

Table 2 The coefficients a_c , b_c , and c_c , used to calculate C_C for all chambers. When c_c is not meaningful, it is excluded. Additionally, an example of C_C and associated uncertainty, u_c ($k=1$) is given for a measured solution with $\rho = 1.066 \text{ g}\cdot\text{mL}^{-1}$ at $20 \text{ }^\circ\text{C}$ and $[Co] = 1.0 \text{ mg}\cdot\text{g}^{-1}$, both known to $\pm 0.25 \text{ } \%$.

Chamber	a_c	b_c	c_c	C_C	u_c
CRC-12	1.0620	-0.0611	---	1.0031	0.0034
CRC-15R	1.0571	-0.0566	---	1.0032	0.0019
CRC-35R	1.0522	-0.0508	-0.00036	1.0023	0.0044
CRC-25PET	1.0487	-0.0488	---	1.0033	0.0025
CRC-1.8atm	1.0543	-0.0541	---	1.0034	0.0013
AL-400	1.0674	-0.0668	-0.00143	1.0053	0.0011
AL-500	1.0467	-0.0451	---	1.0013	0.0024
ICA	1.0577	-0.0567	-0.00009	1.0028	0.0006
VIC	1.0645	-0.0628	---	1.0024	0.0009