

Analysis of a Radioisotope Calibrator

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The analysis of a radioisotope calibrator is presented. The method used to determine the sensitivity of a detector as a function of photon energy is described. By the use of the sensitivity curve, the response of the radiation detector to any radioisotope can be calculated if the decay data are known.

The need for a versatile, reliable radioisotope calibrator with a high stability and a wide, useful activity range in radiopharmacy and nuclear medicine departments is well recognized (1, 2). The primary purpose of a radioisotope calibrator in the nuclear medicine facility is to assist in the delivery of good patient care by assuring reliable and accurate measurements of the radiopharmaceutical dosages before administration to patients.

Recently, assessments of the comparative performance and accuracy of several radioisotope calibration devices have been reported by several authors (3-9). Some limitations in the calibrator use have been noted. Hauser (4), utilizing results from a group of 33 participants, reported that sources of discrepancy for calibrators arise from discrepancies in the assayed activity of commercial radiopharmaceuticals and from poor calibration or insufficient information from manufacturers of certain calibrators. Hare et al. (5) evaluated calibrators in 14 institutions and frequently found 15-25% differences between Ge(Li) detector-calibrated activities and activity values measured by calibrators. The measurement errors were the largest for calibrators with analog meters. Payne et al. (6) found that calibrators manufactured by 11 different firms were accurate to $\pm 5\%$ for ^{99m}Tc , but lacked agreement up to $\pm 100\%$ for the assay of ^{133}Xe . Lundehn (7) has tested eight commercially available calibrators in Sweden and reported that, as of Oct. 1974, none of the calibrators he tested were absolutely calibrated for all source configurations, volumes, etc., although he too noted considerable variation in accuracy between various suppliers. Lundehn concluded that "The best results were obtained with a tall, big ionization chamber with pressure. The best conditions of radiation protection are obtained with lead around the ionization chamber or by a separate ionization chamber and electronic unit." Neirinck et al. (8) and

Johnson et al. (9) have reported that dose calibrator readings may be seriously affected by radionuclidic impurities such as the presence of ^{124}I impurity in ^{123}I .

In view of the difficulties which have been observed in the assay of certain radioisotopes (e.g., ^{133}Xe , ^{125}I , ^{123}I), we have initiated a systematic evaluation of calibrations of radioisotope calibrators. The method used to determine the sensitivity curve of a detector is presented.

Once the curve of the sensitivity of the detector as a function of photon energy is established, the response of the detector to any radioisotope may be calculated, provided that the decay data are known. The required feedback ratio for an amplifier, attenuation ratio, or calibration number needed to give a direct reading of the activity on a meter of a calibrator can hence be found, i.e., plug-in modules or a discrete gain setting switch can be designed or calibration numbers for a continuously adjustable potentiometer can be determined.

This calibration method is applicable to a calibrator with any type of detector. An example of analysis for a calibrator with an ionization chamber is presented in some detail. It is not within the scope of the present studies to determine the sensitivity curve and the accuracy of all types of calibrators. R. Ayres of the National Bureau of Standards (NBS) has recently initiated such comparative studies (10).

Methods and Materials

Definition of response and sensitivity. The response R of the detector to a radioisotope A is defined as the ratio of the detector output to the activity of the radioisotope being measured. It is very convenient to express the response relative to that of the reference standard radioisotope.

$$R_A \equiv \frac{\text{detector output due to sample A}}{\text{activity of sample A}} \div \frac{\text{detector output due to reference source}}{\text{certified activity of the reference source}} \quad (1a)$$

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Cobalt-60 was selected as the reference standard radioisotope of the response studies for the following reasons.

1. Its decay scheme is relatively simple and well established.
2. It is one of the most commonly used radioisotope standards.

Therefore,

$$R_A \equiv \frac{\frac{\text{detector output due to sample A}}{\text{activity of sample A}}}{\frac{\text{detector output due to standard } ^{60}\text{Co}}{\text{certified activity of the standard } ^{60}\text{Co}}} \quad (1b)$$

The sensitivity of the detector for photon energy of E_γ is defined as the detector output due to 3.70×10^{10} photons of energy E_γ , and it is expressed relative to the output of the detector due to unit activity (1 Ci) of reference radioisotope, i.e., ^{60}Co :

$$S(E_\gamma) \equiv \frac{\text{detector output due to } 3.70 \times 10^{10} \text{ photons of } E_\gamma}{\text{detector output due to 1 Ci of } ^{60}\text{Co}} \quad (2)$$

The detector response R_A to radioisotope A, defined by Eq. 1b, and the detector sensitivity defined by Eq. 2 have the following relation:

$$R_A = \sum_i I_i S_i, \quad (3)$$

where I_i and S_i are, respectively, the intensity (mean number of photons per nuclear transformation) and the detector sensitivity corresponding to the photon radiation of energy E_i from the isotope of interest. The response and the sensitivity have the same numerical value if the source of interest decays with monoenergetic photon emission of 100% intensity.

The procedure is to measure the response of the detector (calibrator) to all the available standard radioactive sources as accurately as possible and to establish the sensitivity of the detector as a function of the photon energy so as to satisfy Eq. 3 for all of the standard samples. Once the sensitivity curve has been determined, the chamber response to a radioisotope may be calculated using Eq. 3.

Standard materials. A systematic evaluation of the radioisotope calibrator responses and sensitivity studies was made possible by the availability of a large number of new radioisotope standard materials, especially several low-energy gamma and/or x-ray emission dominant sources.

Radioactive standards certified by NBS, Washington, DC, or by the Laboratoire de Metrologie de la Radioactivit  (LMR), Gif-Sur-Yvette, France, were used for the present work.

The certified standards used for the response studies are listed below together with the reported total uncertainty in their activity. All of the NBS standards, with the exception of ^{133}Xe , were of the liquid

solution form. Approximately 5 g of radioactive liquid were sealed in borosilicate glass ampoules (11) having a diameter of about 1.7 cm, a length of 4 cm, and a wall thickness of 0.06 cm. The ^{133}Xe standard was sealed together with inactive xenon gas in a borosilicate glass ampoule having a volume of about 5 ml, a length of 4.5 cm, a diameter of 1.5 cm, and a wall thickness of 0.13 cm.

NBS certified standards: ^{22}Na (1.7%), ^{51}Cr (1.25%), ^{57}Co (1.7%), ^{60}Co (1.0%), ^{85}Sr (1.3%), ^{110m}Ag (0.7%), ^{113}Sn · ^{113m}In (2.7%), ^{125}I (2.0%), ^{133}Xe (1.8%), ^{134}Cs (2.3%), ^{137}Cs · ^{137m}Ba (2.0%), ^{139}Ce (2.0%), ^{144}Ce ^{144}Pr (2.8%), ^{203}Hg (1.1%), ^{207}Bi (1.7%), and ^{226}Ra + chain of daughters (0.5%).

LMR certified standards— ^{60}Co (1.5%), ^{137}Cs ^{137m}Ba (2%), and ^{241}Am (1%)—were also similarly enclosed in ampoules, except that the solution volume of ^{241}Am was 1 ml.

Sensitivity curve. Ionization chamber responses to the standard sources were measured for a representative number of types of calibrators. The measurements were always made relative to bench-mark standard sources (^{60}Co , ^{57}Co , ^{226}Ra , and/or ^{241}Am) in order to minimize errors due to sensitivity change and/or zero point drift of the electrometer amplifiers.

As a first approximation, the curve of sensitivity as a function of photon energy was drawn assuming that the detector output (e.g., current) arises essentially from the major radiation component of the radioisotope standard samples.

Cobalt-60, the reference standard isotope, was used to determine the first point of the sensitivity curve. (The sensitivity of the ionization chamber was normalized by its response to ^{60}Co as shown in Eq. 1b.) Since the energies of the two photons associated with the beta decay of ^{60}Co (1173 keV, 99.9%; 1333 keV, 100%) are close together, it was assumed, as a first approximation, that ^{60}Co emits two photons of energy 1253 keV (the average energy). Thus, the first approximation for the chamber sensitivity at 1253 keV is 0.50.

The second point of the chamber sensitivity curve was obtained from the measurement of the ^{57}Co standard source. The approximated sensitivity for the average energy of 123.7 keV was found by omitting the contributions from the 14-keV and 692-keV gamma rays.

The third point (60 keV) of the first approximation of the sensitivity curve was obtained from the data for ^{241}Am . The contribution from the 14-keV (29%) x ray was estimated by placing the standard source ampoule in copper cans with various thicknesses and measuring the corresponding chamber output.

The measurement of the ^{125}I standard provided the fourth point, using the average photon energy of 28.4 keV.

The sensitivity approximation for 662 keV was obtained from ^{137}Cs · ^{137m}Ba , using the result from ^{125}I to

subtract the contribution from the 6.9%, 33-keV x ray emitted by ^{137}Ba .

Sodium-22 emits two intense photons: the 1274.5-keV gamma, which is very close to the average energy of the ^{60}Co photons, and the 511-keV annihilation gamma. Thus, ^{22}Na was used to find the sensitivity at 511 keV.

Proceeding in a similar manner, additional points on the sensitivity curve were found. As more points were found, it was possible to upgrade the sensitivity curve successively by breaking up the points representing averaged energies into their component parts and by including the effects of minor radiation components.

All of the available standards were used to determine the sensitivity curve. Unfortunately, there were only a few standards available in the region around 200 keV, where the curve exhibits a local minimum. The region below about 100 keV was carefully studied, since the curve goes through a local maximum and then drops sharply to zero sensitivity

around 13 keV. The low-energy cutoff was determined by the use of the minor photon components of ^{241}Am and ^{57}Co . The study of the local maximum was very important in resolving the discrepancies in the measurement of ^{133}Xe .

Since all the standard sample configurations are similar to each other, no corrections were made for source configuration except for ^{133}Xe . A correction factor of 1.05 was applied to the chamber response for the ^{133}Xe standard to account for the difference in the ampoule thickness and the form of the standard source.

Results

Example. Figure 1 depicts the sensitivity of a typical ionization chamber of a calibrator as a function of photon energy up to 1.9 MeV. Above a photon energy of 200 keV, the ionization in the chamber is mainly due to electrons resulting from Compton scat-

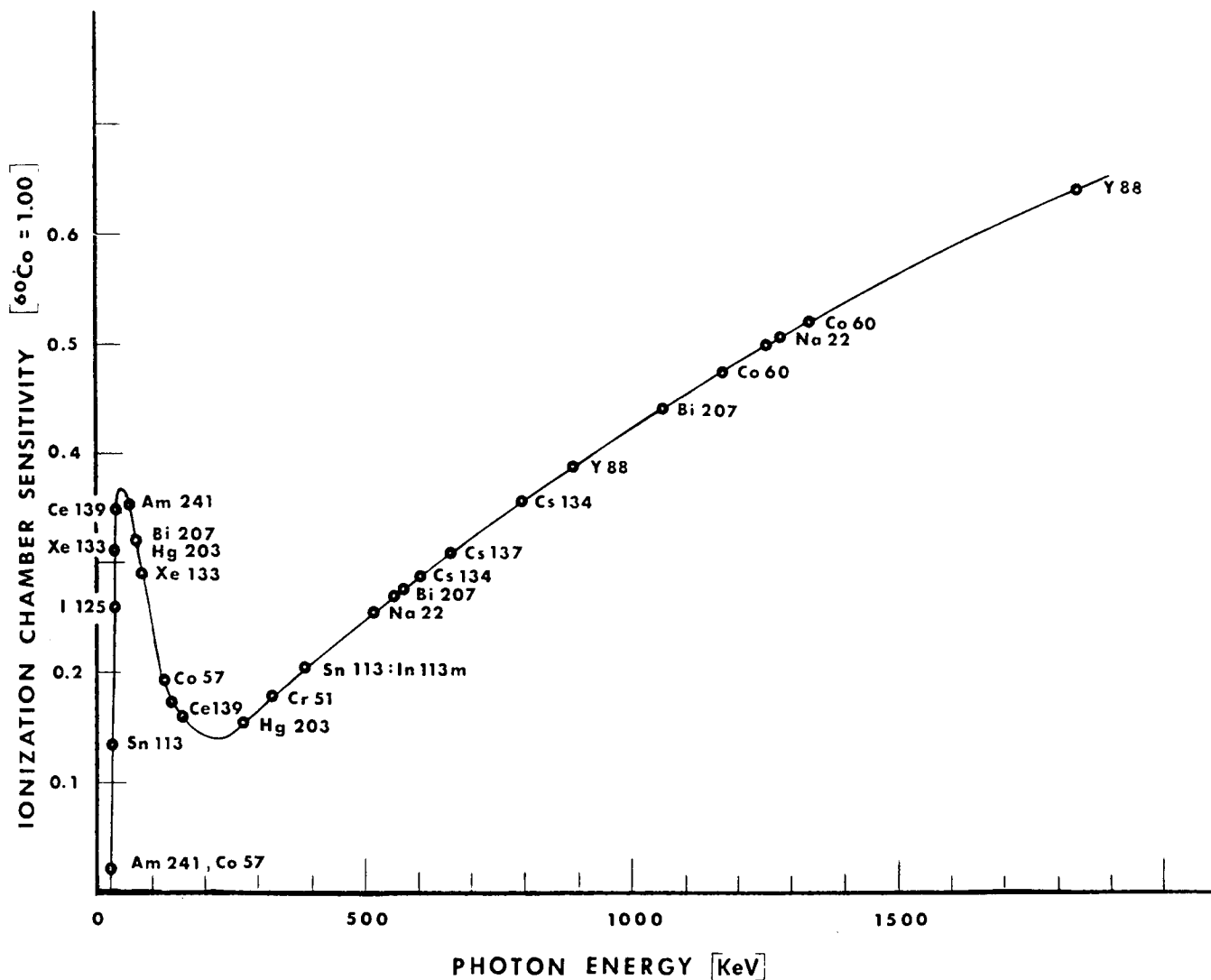


FIG. 1. Sensitivity of typical ionization chamber of calibrator as function of photon energy. Sensitivity is normalized by chamber response to ^{60}Co radiations.

tering of photons by the filling gas (argon) and by the chamber walls (aluminum). The peak in the low-energy region of the sensitivity curve is due to the rapid increase in photoelectric effect as photon energy decreases ($E^{-3}Z^4/A$ per unit mass of the media), and to the attenuation of low-energy photons by the sample holder, the chamber liner, and the chamber walls, as well as the absorption of photons in the sample material and its container.

Although a significant fraction of photons with energies below 50 keV were stopped in the chamber wall, some photons could enter the sensitive volume of the chamber and could, therefore, contribute to the activity measurement. All photons with energies below about 13 keV were stopped before they reached the sensitive volume of the chamber and, therefore, these photons did not contribute to the activity measurement.

At low energies, the response of a particular chamber is extremely dependent upon the type of filling gas and its pressure, the chamber walls and internal electrode materials and their thicknesses, as well as the geometry of the chamber and the external shield configuration.

Calculation of response. After the complete curve of the detector sensitivity versus photon energy is drawn, the response of the radiation detector to any radioisotope can be determined if the energies and the intensities of all gamma and x rays are known. When x-ray data are not found in a reference (12–17), or elsewhere, their intensities arising from internal conversion and electron capture processes must be calculated (18–21). It is also necessary for photons to be the dominant source of detectable radiation.

For each photon energy, the sensitivity is found from the curve. The response of the chamber to a radioisotope can then be calculated using Eq. 3.

Beta-ray correction. The contribution from beta-ray emission may have to be included in the calculation of the response for radioisotopes which are accompanied by high-energy beta-ray emission. It is often difficult to define explicitly a detector's sensitivity to beta rays because of the energy distribution of the emitted beta rays and because of the strong absorption of the radiation by the media. Only bremsstrahlung is detectable by most calibrators due to the rather thick wall of the detector and the sample container.

It is possible, however, to estimate the correction due to high-energy beta rays when these make only a small contribution to the radiation measurement. The contribution from beta rays with a maximum energy of 1.5 MeV or less is totally omitted from the analysis presented here.

The chamber response to a sample of ^{32}P , a pure beta emitter ($E_{\text{max}} = 1.7$ MeV, $E_{\text{av}} = 0.7$ MeV), was measured. The chamber response to a certified stan-

dard source of $^{144}\text{Ce} \cdot ^{144}\text{Pr}$ —which emits an intense, high-energy beta ray ($E_{\text{max}} = 3.0$ MeV, $E_{\text{av}} = 1.24$ MeV) along with several less intense gammas—was also measured. The difference between the measured chamber response to $^{144}\text{Ce} \cdot ^{144}\text{Pr}$ and the calculated response to gamma rays from $^{144}\text{Ce} \cdot ^{144}\text{Pr}$ gives the beta-ray contribution to the activity measurement of the $^{144}\text{Ce} \cdot ^{144}\text{Pr}$ source. Using these two points, ^{32}P and $^{144}\text{Ce} \cdot ^{144}\text{Pr}$ beta rays, the contributions from high-energy beta rays in other radioisotopes were estimated by interpolation of response versus maximum energy. The estimated values of beta-ray contributions were used only as corrections to gamma-ray measurements and no attempt was made to establish the sensitivity curve of the chamber to beta rays.

The gain setting. The gain (or attenuator) of a calibrator amplifier (or output) must be adjusted for each radioisotope in order for the instrument to give a direct reading of the activity of the radioisotope sample. The relationship between the response R_A of the detector and the gain G_A (relative to that for ^{60}Co) is given by

$$G_A = 1/R_A. \quad (4)$$

Even though the practical method of setting the gain for different radioisotopes varies widely from one manufacturer's calibrator to another's, Eq. 4 is applicable to any type of calibrator.

Discussion

Accuracy of the sensitivity curve. The accuracy of the sensitivity curve was tested by calculating the chamber response for all the radioisotope standards used for the present studies. The agreement between all the calculated and the observed responses was within $\pm 3\%$. The accuracy of the chamber-response calculation for a particular radioisotope depends not only on the accuracy of the chamber sensitivity curve but also on the accuracy of the photon intensities given in the nuclear data or the accuracy of the calculation of the x-ray intensity or both.

Effects of an external shield. The advantage of the shield is the reduction of radiation exposure to the personnel handling the radioisotopes, as well as reduction of the background effects on the activity measurements. It is important to note, however, that if a shield is placed around or near a calibrator, the sensitivity of the ionization chamber is enhanced due to backscattering of photons by the shielding. Above about 250 keV the scattering of photons is mainly forward and at the low-energy region; attenuation of photons by the outer wall of the chamber becomes significant. In general, the backscattering effects are more significant for photons of energies between 70 and 250 keV than for photons in other energy regions.

It is not unusual to have an erroneous activity reading of more than 20% if a shield is placed around the

TABLE 1. Summary of Calibration Settings for Shielded CRC-Series Radioisotope Calibrators. Data Derived From Certified Standard Radioisotope Samples and From Published Nuclear Data.

Nuclide	Calibrator setting	Nuclide	Calibrator setting
⁷ Be	179 × 10	⁹⁵ Zr · ^{95m} , ⁹⁵ Nb (total)†	145
¹¹ C	457	⁹⁹ Mo (in canister)	030 × 3.5
¹³ N	457	⁹⁹ Mo (eqb ^{99m} Tc)*	180
¹⁵ O	462	^{99m} Tc	091
¹⁸ F	439	^{99m} Tc (eqb ⁹⁹ Mo)*	195
²² Na	957	⁹⁹ Mo · ^{99m} Tc (total)†	160
²⁴ Na	670 ÷ 2	¹⁰³ Pd	008‡
²⁸ Mg (eqb ²⁸ Al)*	698 ÷ 2	¹⁰³ Ru · ^{103m} Rh	180
²⁸ Al (eqb ²⁸ Mg)*	698 ÷ 2	^{110m} Ag	544 ÷ 2
²⁸ Mg · ²⁸ Al (total)†	698	¹¹¹ Ag	097 × 10‡
³² P	550 × 100‡	¹¹¹ In	331§
⁴⁰ K	555 × 10	^{113m} In	091§
⁴³ K	435	¹¹³ Sn · ^{113m} In	194
⁴⁷ Ca	373	¹²³ I	308
⁴⁷ Sc	036	¹²⁴ I	580§
⁴⁷ Ca · ⁴⁷ Sc (total)†	103	¹²⁴ Sb	720
⁵¹ Cr	100 × 10	¹²⁵ I	319
⁵² Fe	382	¹²⁵ Sb	289
⁵² Mn	676 ÷ 2	¹²⁵ Sb (eqb ^{125m} Te)*	371§
⁵⁴ Mn	309	¹²⁵ Sb · ^{125m} Te (total)†	364§
⁵⁶ Co	648 ÷ 2	^{125m} Te	259
⁵⁷ Co	112	¹²⁶ I	369§
⁵⁸ Co	389	¹³⁰ I	973
⁵⁹ Fe	435	¹³¹ I	146
⁶⁰ Co	990	¹³² I	581 ÷ 2
⁶⁴ Cu	018	¹³³ Xe	188
⁶⁵ Zn	176	¹³³ Ba	555§
⁶⁷ Ga	122	¹³⁴ Cs	726
⁶⁸ Ga	416	¹³⁷ Cs · ^{137m} Ba	220
⁷⁵ Se	230	¹³⁹ Ce	352§
⁷⁶ As	136	¹⁴¹ Ce	066§
⁷⁹ Kr	041‡	¹⁴⁴ Ce · ¹⁴⁴ Pr	387 × 10§
⁸¹ Rb	558	¹⁶⁹ Yb	844§
⁸² Br	541 ÷ 2	¹⁹² Ir	408
⁸⁴ Rb	337	¹⁹⁵ Au	336
^{85m} Kr	074	¹⁹⁷ Hg	232
⁸⁵ Sr	193	¹⁹⁸ Au	143
⁸⁶ Rb	394 × 10	¹⁹⁹ Au	193
^{87m} Sr	095	²⁰¹ Tl	318§
⁸⁷ Y	170	²⁰³ Hg	093
⁸⁷ Y (eqb ^{87m} Sr)*	357	²⁰³ Pb	356
⁸⁷ Y · ^{87m} Sr (total)†	341	²⁰⁴ Tl	420 × 100
⁸⁸ Y	465 ÷ 2	²⁰⁷ Bi	846
⁹⁵ Nb	285	²²⁶ Ra (+ chain)	778¶
⁹⁵ Zr	271	²⁴¹ Am	055§

*eqb represents parent and daughter in equilibrium.

†total represents total activity of both nuclides.

‡Caution: extremely sensitive to assay conditions owing to beta-ray radiations or very low-energy photons; or estimated uncertainty in nuclear data exceeds 20%.

§Container configuration correction may be required; see text.

||Source configuration correction may be required; see text.

¶Read in grams of radium rather than curies. If radium needle (0.5-mm point) is measured, meter reading will be about 10% lower than true value owing to shielding effects of needle.

ionization chamber of a calibrator which was originally calibrated without a shield or when the calibrator is used without the shield if it was originally calibrated with a shield.

Effects of container. The radioactive standard materials in the ampoules now being provided by NBS are a good approximation to an assay of a radiopharmaceutical in a plastic syringe or in a glass syringe (a wall thickness of about 1.2 mm), even for radioisotopes which decay with a significant abundance of

low-energy photons. The user should select, wherever possible, a standardized procedure, volume, and container for all radioactivity measurements. The plastic syringe is convenient since it represents the delivery vehicle to the patient in most clinical situations. Significant errors will occur in some instances, e.g., if the radioisotope is assayed in an appreciably different material and/or wall thickness than that of the standards. The ampoules (11) of recently available standards from NBS are relatively uniform. Plastic sy-

ringes also have a rather uniform wall thickness and absorption is low. However, a random sampling of 5-, 10-, 25-, 50-, and 125-ml-size multiinjection dose vials from several sources indicated that the wall thickness varied randomly from 1 to 3 mm quite independently of the volume of the glass vial.

The assay of radioisotopes having a significant abundance of low-energy gamma-, x-, and/or high-energy beta-ray radiation may be affected by changes in the sample configuration used to assay the radiopharmaceutical, if the procedures are severely different from those recommended by the manufacturer of a given instrument. In such cases, an independent check or determination of a calibration appropriate to a user's particular needs is advised. Fortunately, most radioisotopes can be accurately assayed independently of the sample size if the ionization chamber well is much larger than the sample size and the sample is placed in the center of the well.

The radioisotopes most sensitive to source configuration and type of container are ^{125}I and ^{133}Xe . Other radioisotopes which fall into this category are ^{123}I , ^{169}Yb , ^{201}Tl , and other radioisotopes which decay with significant low-energy photon emission. It is not unusual to have a required correction factor of 2 if ^{125}I is measured in a glass vial.

Effects of impurities. An ionization chamber itself does not have intrinsic energy-discrimination capability. The presence of radioisotope impurities will affect the reading of the instrument (1, 8, 9) unless the effect of impurities is eliminated by photon filtration as is done with ^{99}Mo breakthrough in $^{99\text{m}}\text{Tc}$. However, the presence of a low-level radionuclidic impurity does not negate the usefulness of a radioisotope calibrator, if the user is aware of its presence and has an independently determined calibration including photons arising from the impurities.

In order to assure the accuracy and the linearity of the calibrator at all times, the response of any radioisotope calibrator in clinical use should be checked with 0.1–1-mCi calibration sources such as ^{57}Co (124 keV), ^{137}Cs (662 keV), and ^{60}Co (1.17, 1.33 MeV). The calibrators should also be checked periodically for saturation of the detector. It can be easily tested by measuring the activities of the whole and of a fraction of a high-activity sample and comparing the measured activities. The results of these tests should be recorded in a log book so that changes in the response may be predicted. Tests are also of value in determining the cause of any trouble which may arise.

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