Effects of Characteristic X-Rays on Assay of I-123 by Dose Calibrator

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Large differences in dose-calibrator readings are obtained if "high-purity" I-123 is assayed in different containers. Large correction factors are necessary for assaying another isotope of iodine, I-125, in a dose calibrator, because of absorption of the low-energy (28.4-keV, weighted mean) emissions. We found that up to 70% of the dose-calibrator response to I-123 can be due to characteristic x-rays with energies exactly the same as those emitted by I-125, and that dose-calibrator response to I-123 is also strongly affected by the absorption properties of the vial. An appropriate method to define I-123 activity uses a gamma camera with a medium-energy collimator to establish correction factors for dose-calibrator assay of I-123 in different containers. Correction factors for a plastic syringe and a thick-wall glass vial, were determined using this method. Measurement of I-123 activity in a copper absorber will eliminate the response to x-rays, and the gamma camera is useful in establishing the necessary correction factors.

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In the process of comparing gamma-camera sensitivities to Tc-99m and I-123, as prepared by the indirect reaction, I-127 (*p*,5*n*) Xe-123, we encountered serious disparities between measurements when a dose calibrator was used to define I-123 activity. Typical measurements of I-123 in plastic syringes with a medium-energy collimator gave (per "dose-calibrator unit of activity") only 55–70% of Tc-99m counting rates, instead of the ~91% derived from relative abundance—and efficiency data. This means that the response to I-123 in plastic syringes is higher than normal dose-calibrator response.

It has been demonstrated (*1*) that dose-calibrator response to I-123 may be dominated by the characteristic K x-ray emission, and that there is need to apply correction factors for different source containers. At our institution a further problem was encountered in measuring I-123 activity when the supplier began using different vials, which introduced uncertainty into all our I-123 activity measurements. We decided to develop some method of defining I-123 activity to the extent necessary to establish correction factors for dose-calibrator assay with different source containers.

MATERIALS AND METHODS

To provide a temporary working standard, an I-123 millicurie was defined as "the activity that produces 0.91 of the counting rate of one millicurie of Tc-99m (by dose calibrator) when measured on a gamma camera (159 keV for I-123 and 140.5 keV for Tc-99m, 20% window), with a medium-energy collimator, and with a NaI(Tl) crystal at least $\frac{3}{8}$ in. thick." The factor 0.91 is derived from relative photon abundances and the crystal's efficiency data: (0.834 abundance × 0.90 efficiency)/(0.8907 × 0.925), (*2*,*3*, Appendix). Sources were imaged at 15 cm from the center of the collimator, which was specified as "medium-energy" to avoid the penetration incurred at 159 keV by collimators designed for 140.5 keV. A standard large-field camera was used.*

A sample of high-purity (I-127 (*p*,5*n*) Xe-123) I-123 was obtained from Crocker Nuclear Laboratory,† packaged in the same type of vial (5 ml in flame-sealed ampoule, 0.6-mm walls) used by the Radioactivity Section, NBS, and used in the calibration procedure (*4*) for our dose calibrator‡. Iodine-125 was the only significant contaminant indicated by the supplier's radionuclidic purity data for this product; it was stated to be "less than 1.4%" at 24 hr after time of calibration (TOC).

To determine experimentally the relative contributions of x-rays and gamma photons to the dose calibrator's response, three sources were assayed: bare, and in copper cans 0.010 and 0.020 in. thick. These nine measurements with different absorbers provided three sets of simultaneous equations, which were solved for relative responses to x-rays, 159-keV photons, and higher-energy gammas (184–784 keV, see "other" in Table 2). The sources are identified in Table 2, and a typical set of measurements is shown in the Appendix.

Attenuation characteristics of the copper cans for these measurements were determined in the dose calibrator using I-125 (as a stand-in for I-123 x-rays) and Tc-99m. Figure 1 compares the

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TABLE 1. COMPARISON OF ACTUAL ATTENUATIONS ACROSS COPPER ABSORBERS AND GLASS VIALS IN DOSE-CALIBRATOR MEASUREMENTS OF I-125 AND Tc-99m, WITH NARROW-BEAM ATTENUATIONS

	Transmission factor, I/Io			
		Source container		
Absorber	Narrow-beam	NBS ampoule	10-ml vial	Plastic syringe
For I-125 (28.4 keV)				
0.005-in. copper	0.247	0.191	0.191	0.17
0.010-in. copper	0.103	0.042	0.045	0.036
0.020-in. copper	0.006	0.0038	0.0039	0.0031
0.6-mm glass	0.935	—	—	0.877
1.6-mm glass	0.735	—	—	0.556
For Tc-99m (140.5 keV)				
0.010-in. copper	0.97	0.906	0.913	0.921
0.020-in. copper	0.933	0.84	0.856	0.874

decay schemes and emission spectra of I-123 and I-125 (*5*). The values determined with Tc-99m were adjusted for use with I-123 by multiplying by the measured ratio of the broad-beam attenuation factors determined with a NaI(Tl) detector at 140.5 and 159 keV. For the "other" gamma photons, principally 529 and 539 keV (total 13%), estimated values were derived from mass absorption data.

A NaI(Tl) detector was used with copper foils to determine x-to-gamma ratios [(16–40 keV)/(143–175 keV)] as a function of time (21 hr), to determine the effect of relative in-growth of the longer-lived I-125 radiocontaminant (if any). This apparatus was also used to measure broad- and narrow-beam attenuation factors for the copper foils (used in the copper cans) and glass vials.

The material as received from the supplier was assayed by dose calibrator, then imaged with a gamma camera by its 159-keV emissions. Iodine-123 solution was then removed from the supplier's vial and transferred to (a) a plastic syringe, (b) a thin-wall glass vial similar to the NBS ampoule, and (c) a 10-ml glass vial ("empty sterile" vial, crimp-top stopper, wall thickness 1.3–1.6 mm), and assayed again by imaging and by dose calibrator. As imaging of 159-keV photons is only slightly affected by container walls, imaging data, corrected as needed by narrow-beam attenuation factors, were used to assess the transferred fractions.

A low-activity sample was made by a series of precision dilutions and measured with a GeLi detector and multichannel pulse-height analyzer for comparison with an NBS-traceable multiradionuclide standard[§].

RESULTS AND DISCUSSION

The derivation of fractional dose-calibrator response due to characteristic x-rays was done to improve understanding of how various containers modify the emergent photon population, and—in the end—led only indirectly to the objective, development of some correction scheme. However, in defining attenuation factors for the copper cans, it became evident that it would be difficult to calculate the factors because of the extremely broad-beam geometry for a source in the dose-calibrator well. In such a geometry, many photons arrive at the detector after oblique paths through the wall of the source vessel, which make the wall effectively thicker. The increase in attenuation is easier to measure than to calculate.

Calculation of attenuation coefficients at 159 keV is further complicated by the fact that in both copper and glass, Compton scattering contributes heavily to total interaction probability.

Determination of attenuation factors for each type of source container was necessary, because the total response of the dose calibrator changed with differing container attenuation of the x-rays.

Table 1 shows attenuation factors for three source containers, with surrounding shields of copper or glass, compared with narrow-beam attenuation factors for the same materials.

An analysis (see Appendix)—using I-123 emission characteristics and a typical efficiency curve for the dose calibrator (*4*) (based on 5-ml source in NBS ampoule)—showed that about 61% of the response to I-123 was due to characteristic x-rays (weighted mean 28.4 keV, 0.869 photon per nuclear transition). Table 2 shows the percent response of the dose calibrator to three hypothesized components of I-123 emissions (x-rays, 159 keV, and "others"), calculated from copper-can measurements with I-123 in various containers.

The fractional responses of the dose calibrator to x-ray components of I-123 emission calculated from measurements (Table 2) are in close agreement with those calculated from theory for the NBS ampoule. This agreement is probably due to use of measured attenuation factors for the copper cans using I-125 sources, although some slight error is introduced by the 7% 35.4-keV gamma emission from I-125 (Fig. 1). Conversely, calculated responses to the gamma emission are not as accurate because the results were very sensitive to small changes in estimated attenuation factors for copper at 159 keV and higher.

The fractional responses to x-rays offered here are specific to the dose calibrator and the sources, and may be different for other instruments (*1*).

GeLi-detector comparison of a precision-dilution sample with a multienergy standard indicated an activity in agreement with the initial dose-calibrator assay at time of calibration (TOC) of the entire sample in the NBS ampoule. The establishment of actual I-123 activity was a problem that transcended the variability due to source vessel.

At 24 hr after (TOC, the 5-ml NBS ampoule source gave a dose-calibrator indication of activity 5.6% higher than "true activity" [as defined by imaging relative to Tc-99m at the same time, which was the same as the activity at TOC corrected with a half-life of 13.221 hr (*2*)]. This could have been due partly to relative ingrowth of I-125. If maximum I-125 contamination were present (1.4% at 24 hr after TOC, as specified by the supplier), the ratio

TABLE 2. PERCENTAGE OF TOTAL DOSE-CALIBRATOR* RESPONSE BY EMISSION COMPONENT

Source	Percent of total response		
	x-rays	159 keV	other
Calculated from theory:			
5 ml, NBS ampoule	60.9	37.6	1.5
Calculated from measurement:			
5 ml, NBS ampoule	60.2	31.0	8.8
1.5 ml in 3 ml plastic syringe	69.9	28.7	1.4
5 ml in 10 ml glass vial	47.0	50.4	2.6

* See Footnotes.

TABLE 3. CORRECTION FACTOR, RELATIVE TO CORRECTED READING OF 5-ml SOURCE, NBS AMPOULE

Source	Multiply reading by factor
5 ml in thin-walled, screw-top vial	1.0
1.5 ml in 3-ml plastic syringe	0.72
5 ml in 10-ml glass vial	1.3

for x-rays to 159-keV gammas would have increased by about 2.3% over the first 24 hr. This would have caused a reading falsely high by about 1.4%. The measured change in x-ray-to-gamma ratio over 21 hr (from 3 to 24 hr after TOC) was 4.7%, accounting for the higher dose calibrator reading but indicating actual I-125 relative ingrowth greater than stated limits of I-125 contamination and ingrowth (*6*). Any remaining discrepancy may reflect acceptable error in the measurement of Tc-99m activity, on which assay by imaging depends.

Correction factors for three source configurations are shown in Table 3, these being based on activity as defined by imaging rather than on the response to the NBS ampoule. It is emphasized that these values are highly specific for the named dose calibrator and containers, and small physical differences can result in large changes in the factors.

The use of copper to minimize the response to characteristic x-rays (*1*) is an elegant concept and is applicable to several radionuclides. With I-123, it eliminates alteration of response due to ingrowth of I-125, and sharply reduces variations due to container attenuation. However, the dose-calibrator response is reduced to about 40% with 0.020-in. (0.5 mm) copper (see Appendix). Defining activity with a gamma camera will be useful in establishing correction factors for the copper.

We believe that use of the gamma camera to define I-123 activity, relative to Tc-99m, is a practical way to establish correction factors for the dose-calibrator assay of I-123 in syringes and vials in common use. This has the definite weakness of being dependent on dose-calibrator accuracy for Tc-99m. It has the additional disadvantage that the comparison must be made at low activity (1 mCi or less) to avoid perturbation of results by excessively different deadtimes, correction for which may be difficult. However, camera deadtime differences between I-123 and Tc-99m may be reduced by eliminating the x-rays with copper (and correcting the counts by different copper attenuations at 140.5 and 159 keV). Definition of activity by imaging has the advantage of furnishing activity of the source in the container for which a correction factor is desired, and need only be determined once for that container. It is energy-selective, and except for deadtime effects, is unaffected

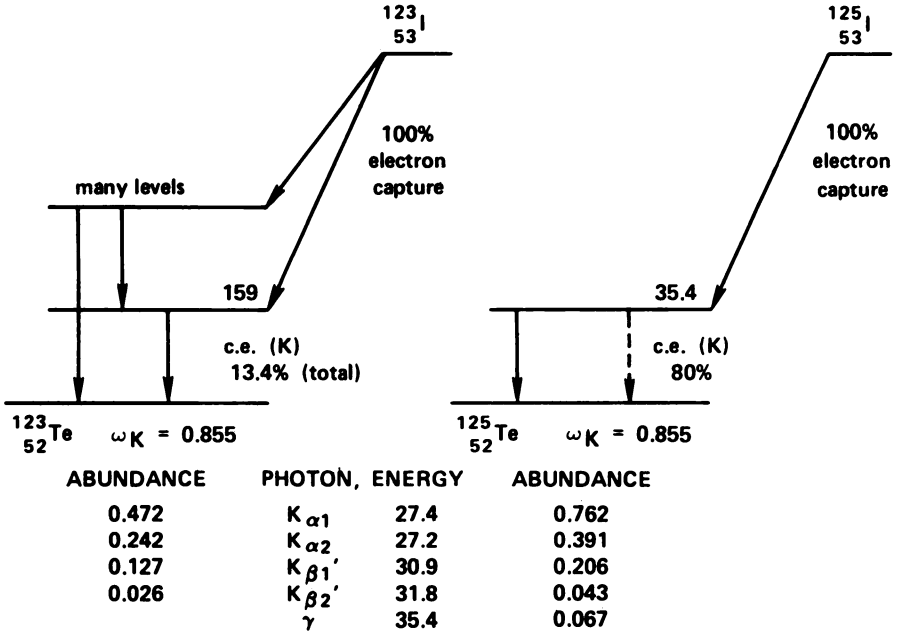

FIG. 1. Simplified decay schemes for I-123 and I-125, from MIRD Pamphlet 10 (*5*). Both electron capture and internal conversion (c.e.) lead to K-shell vacancies; ω_K, fluorescent yield, gives K x-ray photons per K-shell vacancy. Energies are in keV; abundances are in photons per departure from highest energy state. Characteristic x-ray energies are same for both, since proton number of emitting atom (Te) is same for both.

by characteristic x-ray emission. It is also useful in the derivation of correction factors to be applied if copper filters are used to eliminate x-ray response (*1*).

We recommend that some method of correction for the influence of characteristic x-ray emission by I-123 be used in dose-calibrator measurements. If only two source containers are normally used, simple correction factors may suffice. However, there may be no such thing as a standard "empty, sterile 10-ml vial." Therefore the use of copper attenuating cans (*1*), with walls 0.5 to 1 mm thick, may be indicated. In any event, comparison imaging with Tc-99m, with a medium-energy collimator and at low counting rate, provides a means of establishing the necessary correction factors.

FOOTNOTES

* ZLC 37, Siemens Medical Systems, Inc., Iselin, NJ.

† The University of California, Davis.

‡ Model CRC-5, Capintec, Inc., 540 Alpha Dr., Pittsburgh, PA 15238.

§ Mixed radionuclide gamma-ray reference standard QCY44, No. Z20171, Amersham Corp., 2636 S. Clearbrook Dr., Arlington Heights, IL 60005.

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APPENDIX

Gamma photon abundances. The following are gamma and x-ray abundances from Refs. *2*, *5* and *7* (for relative counting rates those from Ref. *2* were chosen because they are the latest):

	Tc-99m 140.5 keV	I-123 159 keV	I-123 x-rays	I-125 x-rays & 35.5 keV
Kocher (*2*)	0.8907	0.834	0.869	1.43
MIRD 10(*5*)	0.8787	0.836	0.867	1.467
NCRP 58 (*7*)	0.8897	0.829	0.866	—

All of the above have published uncertainties ranging from 1% to 5%. The peak efficiencies assumed from Ref. (*3*)—0.925 for 140.5 keV and 0.9 for 159 keV—are subject to error. The calculated ratio of 0.91 for I-123 to Tc-99m counting rates is therefore approximate.

Sample calculation for fractional response. In the equation below, R_x is the fractional response to x-rays, R_g is the response to the 159-keV gamma, and R_0 the response to "others." The numerical coefficients are copper transmission factors at the component energy for the named source.

Typical measurement in copper cans, with the resulting equations, for 5 ml in NBS ampoule:

	(reading)		(x-ray)		(159 keV)		("other")
bare:	7.298	=	$1.0\ R_x$	+	$1.0\ R_g$	+	$1.0\ R_0$
0.01-in:	3.195	=	$0.042\ R_x$	+	$0.934\ R_g$	+	$0.9995\ R_0$
0.020-in:	2.893	=	$0.0038\ R_x$	+	$0.885\ R_g$	+	$0.999\ R_0$

The three equations with three unknowns are solved, with the following result: $R_x = 4.393$, $R_g = 2.262$, $R_0 = 0.642$. The percentage responses, expressed as percentages of 7.298 (the bare indication) are: $R_x = 60.2\%$, $R_g = 31\%$, and $R_0 = 8.8\%$ (Table 2).

Analysis of theoretical fractional response. The following is an approximate analysis of calibrator & response based on MIRD 10 (*5*) abundances and a typical published efficiency curve for 5-ml sources in NBS ampoule (4):

Component	Abundance		Efficiency (arbitrary)		Rel. Resp	% Resp
x-rays	0.8671	×	0.257	=	0.223	60.9
159 keV	0.833	×	0.165	=	0.137	37.6
184-784 keV	0.231	×	0.239	=	0.0055	1.45

The efficiency for 184-784 keV is a weighted average.

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