DETERMINATION OF THE LIQUID SCINTILLATION COUNTING INEFFICIENCY (WALL EFFECT) FOR ALPHA EMITTERS, USING THE ALPHA-GAMMA ANTI-COINCIDENCE METHOD

Ryan Fitzgerald[1] • Anne Marie Forney

National Institute of Standards and Technology, MS8462, 100 Bureau Drive, Gaithersburg, Maryland 20899, USA.

ABSTRACT. The wall effect in liquid scintillation counting (LSC) is the loss of efficiency in the case that an α particle hits a surface (wall or air) before depositing enough energy to be detected. We report our measurements of this LS inefficiency using the 4πα-γ anticoincidence method with corrections for the presence of γ-rays, X-rays, and electrons during some α decays. We derive the Benjamin equation and test the application of this equation to LS α counting. Our value for the LS inefficiency is $(6 \pm 5) \times 10^{-5}$ for a typical low-energy threshold level. This value is consistent with most literature values, but is smaller than the value reported by Cassette (2002) of about 2×10^{-3}. We discuss possible reasons for this disagreement.

INTRODUCTION

Basson and Steyn (1954) first demonstrated the measurement of α activity by liquid scintillation counting (LSC). Early work on the method then focused on developing stable LS cocktails and improving scintillation efficiency and energy resolution. Meanwhile, Engelkemeir and Libby (1950) had already characterized the "wall effect" for internal gas proportional counting (in addition to the end-field effect). That is, β particles are not counted if they hit a wall before depositing enough energy in the detector volume to pass a threshold.

Horrocks and Studier (1958) developed an LS cocktail with reduced chemical quenching and improved energy resolution for counting mixtures of α emitters and β emitters. The α peak tailing that they observed was small enough that the portion of a ^{239}Pu α spectrum that was within the ^{241}Pu beta region (endpoint 21 keV) was only about 0.1%. The α inefficiency was thus less than about 0.1% at their typical noise threshold of about 5 keV. In a 1965 international comparison of ^{241}Am activity determinations (Rytz 1963), the average activity reported for 4 non-wall-effect-corrected LS measurements was 0.5 ± 0.4% higher than the average for 15 α-γ coincidence determinations, indicating no wall effect at that uncertainty level (Brouns 1968). Ihle et al. (1967) compared unquenched ^{233}U LS samples to strongly quenched samples, and saw no counting difference within their 0.1% uncertainty. Vaninbroukx and Spernol (1965) reported ^{241}Am activity ratios from 3 methods relative to LS counting. For 4π proportional counting, 4πα-γ coincidence counting, and low-geometry counting, they report ratios of 1.0012, 1.0000, and 0.9998, respectively, giving an average difference from LS counting of 0.03%. (It is difficult to determine their uncertainties for the individual values, as the paper mostly detailed β counting.) Other wall-effect reports suffered from large uncertainties obscuring the effect.

More recently, Cassette (2002) applied Benjamin's ^{3}H-internal-gas-counting wall effect to LS α counting (stating 4 assumptions), thereby predicting a wall effect of 0.16% to 0.23% for ^{238}Pu. Cassette also made measurements using LS vials into which he had inserted glass plates in order to increase the wall losses. He then extrapolated to zero surface area and determined a loss of about 0.2% for a standard 10-mL LS source using a linear fit of 3 wall-surface values. No uncertainty was assigned to the 3 data points or to the deduced wall effect.

[1]Corresponding author. Email: ryan.fitzgerald@nist.gov.

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In 2007, we measured ^{241}Am two ways: by commercial LS counter and by $4\pi\alpha$(LS)-γ anticoincidence counting at the National Institute of Standards and Technology (NIST). The LS counting activity value differed from the anticoincidence extrapolation by (+0.05 ± 0.22)%[2]; thus, no wall effect was revealed at this uncertainty level (NIST 2007).

In this paper, we report the measured LS α inefficiency using live-timed $4\pi\alpha$-γ anticoincidence counting. We make small corrections to the measured LS inefficiency from effects due to the presence of γ-rays, X-rays, and electrons. We derive Benjamin's equation and carefully apply it to the results. We also provide a test of the hypothesis that the observed inefficiency is caused by the wall effect.

The possibility of these small wall-effect losses is important in metrology. For example, in a recent international comparison of ^{241}Am standardizations reported by Ratel and Michotte (2003) the typical reported uncertainty was 0.12%, which may be the same order of magnitude as the wall effect.

DERIVATION OF THE BENJAMIN EQUATION

Benjamin et al. (1962; abbreviated hereafter as *Benjamin*) provided an equation that was stated to be equivalent to a Monte Carlo simulation for the "fraction of counts crossing the boundaries of the counter with energy *E*." Benjamin stated that this equation was accurate to 1%, which is not saying much, since their calculated correction factors differed from 1 by less than 2%. This equation, derived for ^{3}H counting in an internal gas proportional counter, has been misapplied to LS counting in subsequent literature, as will be described here.

We begin by deriving the equation using simple geometry. First, we derive the fraction Z of the volume of a right cylinder, height h and radius r, that is not within a distance δ of a boundary. Let V be the volume of the cylinder, and V' be the volume of the enclosed cylinder with boundaries a distance δ from the boundaries of V. Then, Z is given by

$$Z = \frac{V'}{V} = \frac{\pi(r-\delta)^2(h-2\delta)}{\pi r^2 h} \tag{1}$$

In the cases considered here (alpha LS counting, ^{3}H proportional counting), $0.98 < Z < 1$ and $\delta/r < 0.01$, $\delta/h < 0.01$, so terms of order δ^2/h^2, $\delta^2/(rh)$, and $\delta^3/(hr^2)$ can be ignored. Thus, this fraction can be expressed as

$$Z = 1 - 2\delta\left(\frac{1}{r} + \frac{1}{h}\right) \tag{2}$$

Now we determine the fraction of those particles emitted isotropically from a location within a distance δ of a surface that will traverse a distance less than or equal to δ before hitting that surface. This can be done by averaging the fractional wall-subtended-solid-angle (Ω) from the 2 extreme cases. If the decay happens on the wall itself, $\Omega = 1/2$ of those alphas will hit the wall immediately, while those alphas emitted in the direction opposite the wall will stop within the volume. At the other extreme, if the decay happens a distance δ from a surface, then $\Omega = 0$ of the alphas will hit the wall since the α particle would have to travel exactly perpendicular to the wall in order to hit the

[2]All uncertainties reported here are combined standard uncertainties, which approximate 1 standard deviation and are equivalent to expanded uncertainties with coverage factor, $k = 1$ (BIPM et al. 1995).

wall. On average, 1/4 of the α particles emitted from locations within δ of a surface will hit that surface before traveling δ. (This same value of 1/4 was derived by integration.) Including this solid angle factor of 1/4 in the volume factor, *Z*, results in the expression *F*, for the attenuation of α particles due to the wall effect:

$$F = 1 - \frac{1}{2}\,\delta\left(\frac{1}{r} + \frac{1}{h}\right) \tag{3}$$

Note that 1–*F* is equal to Benjamin's equation (his page 10). Here, we equate δ to $R(E_e,E_T)$ (our notation), where *R* is the "range" traveled by an electron of initial energy E_e to lose enough energy to pass the detection threshold, E_T (2 keV for Benjamin). Understanding this point is important because a high-energy electron will travel farther to lose E_T than will a low-energy electron. The situation will become even more complicated later when we consider LS counting and quenching.

We have reproduced Benjamin's correction factors for their proportional counter system using $\delta = R(E_e, E_T)$ by calculating the average value of *F*, weighted by the ^{3}H spectral shape (Fermi function). Note that this *F*-modified spectrum does not correspond to the actual measured spectrum, in which some higher-energy electrons will be counted at lower energy, but to the original spectrum minus the counting losses at each energy. We calculate $\bar{F} = 0.986 \pm 0.005$, in good agreement with Benjamin's calculated $\bar{F} = 0.984 \pm 0.004$, and his experimental $\bar{F} = 0.981 \pm 0.004$. However, using the full stopping range at each electron energy, $R(E_e, E_e)$, we calculate $\bar{F} = 0.958 \pm 0.008$, in disagreement with Benjamin's data and calculations.

USE OF THE BENJAMIN EQUATION FOR LS COUNTING

The Benjamin equation is frequently referenced in the LS literature, for example in the oft-cited works by Gibson and Gale (1968) and Gibson (1980). There, *R* was taken to be the full range for an electron of initial energy E_e to lose all of its energy, that is, $R(E_e, E_e)$. Interpreted that way, the Benjamin equation "only gives the proportion of particles that encounter the wall" (Vatin 1980) not the reduction in scintillation response, as claimed by Gibson and Gale. This interpretation of Benjamin's *R* gives the wrong wall effect, as described above.

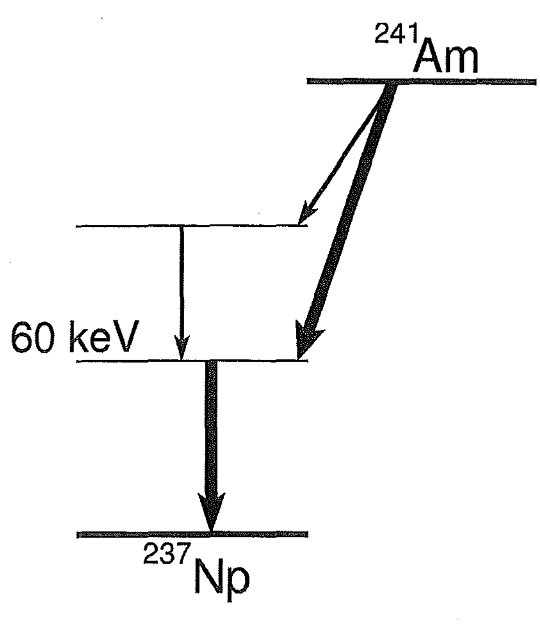

Figure 1 Simplified ^{241}Am α decay scheme. Branching probabilities are listed in the text.

As Cassette (2002) pointed out, application of the Benjamin equation to β LS counting is dubious, due, for instance, to the fact that an LS counter has a nonlinear energy response (due to ionization quenching), unlike a gas proportional counter. Instead, Cassette used a Monte Carlo simulation to determine the modified LS spectra shapes for LS β spectra, and then used those spectra to model the wall effect for typical LS counting methods.

The wall effect is simpler for LS α counting than for LS β counting due to the short range and nearly monotonic spectrum of most α emitters. For ^{238}Pu α decay, Cassette used the Benjamin equation as a rough estimate of the wall effect, after stating some assumptions. He used the full α range, R = 30 μm, in the manner of Gibson and Gale, instead of the range needed for detection, $\delta = R(E_\alpha, E_{\alpha,T})$. He found the alpha inefficiency, i_α, to be about 2×10^{-3}. This result was consistent with his experimental work, in which he added glass plates inside the LS vials to increase the effect, and then extrapolated back to zero surface area.

Note that special care is needed when determining, and even expressing, the LS threshold, due, for one thing, to ionization quenching. For example, if alphas with energy form a peak centered at channel N on a linear spectrum, then channel $N/2$ corresponds neither to an $\frac{E_\alpha}{2}$ alpha losing all its energy, nor to an E_α alpha losing half its energy. This is one of the reasons why an "LS energy calibration" is a tricky task. In this paper, we define an energy threshold, $E_{\alpha,\tau}$, which corresponds to the energy loss of an α particle of initial energy E_α (5.6 MeV for ^{241}Am). Since some LS counters have nominal thresholds defined using electrons, we also "convert" these energies into approximate electron energies, E_e. All of these energy calculations are done using Birks' (1964) equation for ionization quenching. For example, in order for an electron to stop in the LS volume and create the same size LS signal as an E_α alpha that loses $E_{\alpha,T}$ within the LS volume, the electron must have an energy, E_e, that satisfies this equation:

$$\int_0^{E_e} \frac{dE_e'}{1 + k_b S_e(E_e')} = \int_{E_{\alpha,T}}^{E_a} \frac{dE_\alpha'}{1 + k_b S_\alpha(E_\alpha')} \tag{4}$$

where $s(E)$ is stopping power (Berger et al. 2005) and k_b is a quench parameter in units of inverse-stopping power. In the present work, we used k_b = 0.012 cm/MeV and we solved Equation 4 numerically to find equivalent electron thresholds for the alpha thresholds used in the LS counter. We consider these equivalent electron energies to be approximate, since LS counters do not have absolute E_e thresholds. Rather, electrons with energy E_e may produce pulse heights above or below the LS pulse-height threshold. Thus, these electron thresholds are meant only in the sense that they are defined in Equation 4.

USE OF THE BENJAMIN EQUATION IN PRESENT WORK

To summarize our adaptation of the Benjamin equation, we use the following form for the present experimental work:

$$F = 1 - \frac{1}{2}R\,(E_\alpha, E_{a,T})\left(\frac{1}{r} + \frac{1}{h}\right) \tag{5}$$

where $E_{\alpha,T}$ is the LS threshold for an alpha emitted with energy E_α. This equation can be written in terms of the alpha inefficiency, i_α, assuming that the inefficiency is only due to the wall effect, as,

$$F = 1 - i_\alpha \tag{6}$$

METHOD

One goal of this experimental work was to measure the LS α inefficiency, i_α, using the live-timed 4πα-γ anticoincidence method (LTAC) and ^{241}Am LS sources. To do so, we carefully designed the experiment, and then made small corrections to the observables, such that the inefficiency measured in the experiment, i_{exp}, could be used to determine the true i_α.

Figure 1 shows a simplified illustration of ^{241}Am α decay ($E_\alpha \approx 5.6$ MeV). All nuclear data are from the updated *Table of Nuclides* (Chechev and Kuzmenko 2010) and uncertainties are only listed where important for final results. The ^{241}Am decays directly to the 60-keV state of ^{237}Np with a probability of 0.845, and cascades from all higher-energy states to that 60-keV state with probability 0.142. Given the emission of a 60-keV γ-ray (probability 0.36), the 60-keV state is fed directly with probability $P_d = 0.856 \pm 0.001$ and from a higher state with $P_h = 0.144 \pm 0.001$.

The basic strategy was to gate the γ-ray channel of the LTAC system on the 60-keV full-energy peak. The experimental LS inefficiency was approximated, as usual (Baerg 1981), by the ratio of the anti-coincident γ-rays to the total γ-rays (after correcting for background and decay):

$$i_{exp} = \tilde{N}_{\gamma,A/(N_\gamma)} \tag{7}$$

This experimental inefficiency was not exactly equivalent to the alpha inefficiency, i_α, due to the occasional presence of electrons and X-rays. The majority of those signals were excluded by the experimental design since for the 60-keV γ-ray to be counted, that 60-keV state could not decay by conversion electron. This would not be the case if the X-rays were allowed in the NaI energy window. Nonetheless, 14% of the time that the 60-keV γ-ray was detected, it resulted from a cascade that probably created electrons and X-rays (99% of time). Thus, to a good-enough approximation, i_{exp} is the weighted average of i_α (weighted by P_d) and i_e, the inefficiency for the various combinations of electrons, alphas, γ-rays, and X-rays weighted by P_h. A less significant complication arises from the case in which the 60-keV γ-ray scatters in the LS counter, depositing on average ~6 keV, and is then registered within the full-energy peak in the NaI detector. This possibility was explored both experimentally and by Monte Carlo simulation, as described below. The LS efficiency for this scenario varies from $\varepsilon_{LS,\gamma} = 0.04$ to 0 for the threshold ranges reported here, and so only led to a small correction. Considering our small values of $\varepsilon_{LS,\gamma}$ and very small values of i_α for the thresholds used, we can express our desired measurand, i_α, in terms of our measured quantity, i_{exp}, by this equation:

$$i_\alpha = \frac{i_{exp}}{1 - P_h\varepsilon_e - P_d\varepsilon_{LS,\gamma}} \tag{8}$$

The correction to i_α is smaller than the counting-statistics uncertainty on i_{exp} in all cases. For the lowest threshold results, where the effect is largest, $\varepsilon_e = 1$ and $\varepsilon_{LS,\gamma} = 0.04$, so that $i_\alpha = 1.20 i_{exp}$. For large values of $\varepsilon_{LS,\gamma}$, this equation would blow up because if the Compton-scattered γ-rays or electrons were highly detected in all decays (instead of rarely detected in only 14% of them), then it would take an enormous alpha inefficiency in order to observe any anticoincidences. Eventually, we would have to use a more complicated formula. However, this works fine for our case.

Note that we have ignored any contribution to the LS signal from the Np recoils, which have an energy of about 100 keV. Due to ionization quenching, the recoil will not produce significant amounts of light during stopping (Birks 1964). Also, one goal of this work was to determine the magnitude of inefficiency, i_α, for a hypothetical pure-alpha emitter. Even in that hypothetical case, the recoil nuclei would recoil.

EXPERIMENTAL RESULTS

The LTAC instrument contains a single-phototube LS detector surrounded by a NaI(Tl) well detector. The instrument and method are described by Fitzgerald and Schultz (2008) and references therein. For this work, we measured four ^{241}Am sources and 1 blank. The sources consisted of glass quasi-hemispheres of inner radius 1.47 ± 0.13 cm and a filling height of 0.50 ± 0.03 cm. The liquid consisted of nominally 3 mL of Ultima Gold™ AB[3] LS cocktail, 0.2 mL of ^{241}Am solution in 1 mol/L HNO_3 and 0.02 g of HDEHP complexing agent. Each source contained ~5 kBq of ^{241}Am. The blank was prepared in the same way, except 1 mol/L HNO_3 was substituted for the ^{241}Am solution.

The LTAC system was set to 30 μs shared extending-deadtime. The LS linear-amplifier gain was chosen to first center the LS α peak in the linear spectrum. Then, the gain was increased by a factor of 4 for the LTAC measurements. The approximate energy calibration was determined using this 1-point calibration and Birks' integrals. Anticoincidence data was accumulated for 500 live seconds for each of 6 lower-level discriminator thresholds, T, within each measurement cycle. The LTAC system does not have a separate, live-timed N_γ channel, beyond the anticoincident $N_{\gamma,A}$ channel. Instead, N_γ was determined at the beginning and end of each measurement cycle by using an LS threshold of $T = 10$ V, which is above the saturation peak of the LS amplifier (thus, $N_{LS} = 0$). Also, a non-live-timed "monitor" NaI rate was measured during each $N_{\gamma,A}$ measurement. Since the inherent NaI deadtime was constant during the measurement cycle, the ratios of these monitor rates were used to correct N_γ for any drift during the measurement cycle. No significant drift was observed.

Because the LS inefficiencies were so low, the gross anticoincident NaI rates, $N_{\gamma,A}$, that were used to determine i_{exp} were near background levels. Therefore, we took steps to ensure that these $N_{\gamma,A}$ values were correct. Recall that in the anticoincidence method there is no problem of random coincidences (Baerg 1981). Likewise, we ensured that no coincident γ-rays could be counted as anticoincidences by delaying the NaI logic pulses adequately (5 μs) and checking on an oscilloscope that the NaI pulses fell well within the LS-initiated deadtime. The i_{exp} values did not depend on the LS background, but did depend on the $N_{\gamma,A}$ background. For this reason, we measured the blank 6 times both before and after measuring the four ^{241}Am sources. The between-measurement background standard deviation was consistent with the within-measurement counting statistics ($\sqrt{N}$), as shown in Table 1. Also, as expected, there was no correlation between the LS threshold and background $N_{\gamma,A}$ count rate. In separate experimental work using blanks of various volumes, we discovered that the sample-to-sample $N_{\gamma,A}$ background variation is negligible for the sources used here (unpublished data).

The LS γ-ray efficiency ($\varepsilon_{LS,\gamma}$) was determined using an EGSnrc (Kawrakow 2000) Monte Carlo simulation. We also measured the LS response to an ^{241}Am point-source sealed in polyester tape, mounted above the blank LS source. This experimental efficiency was scaled to account for the different geometry between that point-source and a ^{241}Am solution mixed in to the LS cocktail by using Monte Carlo simulations of both geometries. The pure Monte Carlo simulation gave, for a threshold of $T = 0.30$ V ($E_e \approx 10$ keV), $\varepsilon_{LS,\gamma} = 0.044$. The experimental value, using the Monte Carlo to scale to the correct geometry gave $\varepsilon_{LS,\gamma} = 0.033$. We adopted a value of $\varepsilon_{LS,\gamma} = 0.04 \pm 0.02$. The large uncertainty is due mostly to the uncertainty in our energy calibration, and thus the LS threshold.

[3]Certain commercial equipment, instruments, and materials are identified in this paper to foster understanding. Such identification does not imply recommendation or endorsement by NIST, nor does it imply that the materials and/or equipment are the best available for the purpose.

Table 2 shows the experimental inefficiencies, $i_{\exp}$, and corrected inefficiencies, i_α, for each threshold T. For the lowest threshold that is probably near or above the thresholds on commercial LS counters, $i_\alpha = (6 \pm 5) \times 10^{-5}$. This small inefficiency is negligible even in metrological studies. For the highest threshold studied that could be used when discriminating a low-energy β emitter from an alpha emitter, $i_\alpha = (1.50 \pm 0.11) \times 10^{-3}$ could be significant.

Table 1 Number of gross, live anticoincident NaI counts ($N_{\gamma,A}$) in 500 live seconds for four ^{241}Am sources, measured at the lowest threshold of $T = 0.3$ V. Also, the average $N_{\gamma,A}$ value for a blank, B0, averaged over 6 threshold settings, with the average $\nu(N_{\gamma,A})$ within a single 500-s measurement. The standard deviation (SD) of the blank $N_{\gamma,A}$ values for the 6 settings is shown in the final column and is consistent with the counting statistics within each 500-s measurement, as expected.

Sample	$N_{\gamma,A}$	$\sqrt{N_{\gamma,A}}$	SD ($n = 6$)
B0	111	11	8
B3	134	12	
B4	111	11	
B1	136	12	
B2	124	11	
B0	111	11	10

Table 2 Experimental α-inefficiency results. For a given spectrum threshold, T, Birks' integrals were used to determine the equivalent alpha energy-loss threshold, $E_{\alpha,T}$, and electron energy $E_{e,T}$. The experimental inefficiency at a given threshold is given by $i_{\exp}$, with an uncertainty dominated by anticoincident-γ ($N_{\gamma,A}$) counting statistics. The LS efficiency for Compton-scattered 60-keV γ-rays is given by $\varepsilon_{LS,\gamma}$ and the LS efficiency for conversion electrons, Auger electrons, and X-rays is given by ε_e. The experimental inefficiencies were corrected using Equation 8 to determine the α inefficiencies, i_α.

T (V)	$E_{\alpha,T}$ (keV)	$E_{e,T}$ (keV)	$i_{\exp}$	$\varepsilon_{LS,\gamma}$	ε_e	i_α
0.30	70 ± 12	10	$(5.3 \pm 4.1) \times 10^{-5}$	0.04 ± 0.02	0.9 ± 0.1	$(6 \pm 5) \times 10^{-5}$
2.00	480 ± 30	54	$(7.0 \pm 0.7) \times 10^{-4}$	0.005	0.5 ± 0.5	$(7.6 \pm 1.0) \times 10^{-4}$
4.00	980 ± 50	105	$(1.50 \pm 0.11) \times 10^{-3}$	0	0.001	$(1.50 \pm 0.11) \times 10^{-3}$

WALL EFFECT PARAMETER DETERMINATION

Table 3 shows the results for the Benjamin equation length parameter δ, determined from each i_α value. The energy loss of the ^{241}Am alphas as they traverse those δ values was then calculated using the stopping power to be $E(\delta)$, shown in the final column of Table 3. If the deduced i_α are caused by the wall effect and if the Benjamin equation characterizes that effect, then these energy losses, $E(\delta)$, calculated from δ and stopping powers, should be equivalent to the threshold energies, $E_{\alpha,T}$, determined from the spectrum threshold calibration. In fact, these 2 measures of the energy loss agree within their uncertainties. The biggest relative difference is at the lowest threshold where the correction for other efficiencies is significant and the counting statistics uncertainty is also large.

Table 3 Results from α inefficiency measurements. The $E_{\alpha,T}$ are from Table 2. The α inefficiency at a given threshold is given by i_α. The i_α values were used in Equation 5 to determine the length parameter, δ. Finally, the energy loss, $E_{\alpha,T}(\delta)$ of an alpha traveling distance δ was calculated using the stopping power. The agreement of these calculated energies with the spectrum calibration indicates that the measured inefficiencies are consistent with the Benjamin equation.

$E_{\alpha,T}$ (keV)	i_α	δ (μm)	$E(\delta)$ (keV)
70 ± 12	$(6 \pm 5) \times 10^{-5}$	0.5 ± 0.4	35 ± 27
480 ± 30	$(7.6 \pm 1.0) \times 10^{-4}$	5.7 ± 0.8	430 ± 60
980 ± 50	$(1.50 \pm 0.11) \times 10^{-3}$	11.2 ± 1.0	880 ± 80

DISCUSSION

Adapting the above results to a commercial counter requires adjustments for sample geometry and LS threshold. For a typical LS vial filled with 10 mL of liquid, $r = 1.3$ cm and $h = 2.0$ cm. Therefore, the above value of $i_\alpha = (6 \pm 5) \times 10^{-5}$ for our NIST LTAC quasi-hemispherical vials would be $i'_\alpha = (3 \pm 3) \times 10^{-5}$ for a commercial counter with a similar threshold. This small inefficiency is consistent with most of the literature cited above, but is significantly lower than Cassette's (2002) values of 1.6×10^{-3} to 2.4×10^{-3}. Recall that he calculated those values using the full α range of 30 μm, rather than the range needed to pass an energy threshold. Also, he measured a consistent value of about 2 $\times 10^{-3}$ by adding glass plates inside the LS vial to exacerbate the losses, then extrapolating to zero loss.

In conclusion, the present work and Cassette's work attempted to realize the same *measurand*, but we apparently did not. Perhaps Cassette's instrument had a higher threshold than our lowest setting, or he introduced other sources of inefficiency than just the wall effect when he added the glass plates to his vials. Perhaps we have other sources of efficiency that we did not recognize and account for. Future work could reconcile the discrepancy.

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