Follow-up Multicenter Alpha Counting Comparison

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Abstract— **A follow-up alpha emissivity study was conducted to examine the wide variability observed in previous work that was hypothesized to be due to differences in the pulse height discrimination threshold among participant's equipment. Two samples, one mixed energy and one monoenergetic, were prepared and sequentially circulated to all participants for counting. Analysis of the data demonstrates that only a small portion of the variability is explained by this mechanism. The role of the sample to entrance window gap for some counters was analyzed post hoc using the same data set and may be responsible for a large amount of the variability. The results of this large scale study demonstrate the continuing uncertainty for these measurements and the importance of interpreting their results appropriately when estimating soft error rates.**

***Index Terms*—Reliability, materials, alpha particle, detectors, single event upsets.**

I. Introduction

Soft errors in modern electronic products are an important reliability concern. Low intensity alpha particle emission, either from the radioactive decay of materials used in products or contamination during the manufacturing process, may cause errors in susceptible circuit elements and is a large component of the soft error rate (SER) for circuits operating at ground level [1, 2]. Accelerated test methods to quantify a circuit's susceptibility to alpha particle errors, coupled with accurate measurement of the alpha particle emissions from materials and manufacturing monitor samples may be used to assure that a system will meet its requirements related to soft error robustness. This paper describes an assessment of the accuracy of alpha particle counting methods.

Many elemental isotopes are radioactive and undergo either beta decay or alpha decay. Alpha decay is more common for heavy isotopes such as the members of the ^{238}U or ^{232}Th decay chains. These isotopes and their daughter decay products are quite common throughout the earth's crust and are present at low concentrations in the majority of materials used for electronics production. Because the alpha particle has a range of less than 100 μm in most solid materials it is important that the measurement of alpha particle decay be characterized as the functional emission from a material's surface, rather than as a bulk property of the sample. Sample inhomogeneity may result in different surface emissivity as compared to the value expected from bulk concentrations [3, 4]. Attempting to chemically analyze the isotopes responsible for a sample's alpha emission is further complicated by the extremely low concentrations at parts per trillion or less [5, 6], for most materials. For materials with high specific activity, such as ^{210}Po, the allowed concentration may be as low as 1 part in 10^{18}, clearly beyond the capability of any existing chemical analysis [4].

By convention, materials with alpha particle emissivity between 2 and 50 α/khr/cm^2 are referred to as low alpha (LA) and those with emissivity below 2 α/khr/cm^2 are referred to as ultralow alpha (ULA) materials. Recently, the term Super Ultralow Alpha (SULA) has been proposed for emissions below 0.5 α/khr/cm^2. (In past work, when material emissivity was at much higher levels, the common unit was α/hr/cm^2 rather than α/khr/cm^2. As most modern electronics require emissivity values in the ULA or LA range it is now preferred practice to utilize units of α/khr/cm^2 to prevent confusion.) Measurement of alpha particle emissivity at these levels utilizes special purpose detectors and large area samples of at least 200 cm^2. The most commonly-used detector has been a large format gas proportional counter although suitable ionization counters have recently become available. A Geiger mode counter is impractical for measurement at this range of emissivity [7].

An earlier study [8] sought to compare alpha particle emissivity measurements of LA and ULA materials with 9 different participating centers. Two matched LA and two matched ULA samples were prepared and sent to each center for measurement using their own protocols and measurement equipment. Counting data were confidentially submitted for comparative analysis once all measurements were complete. The resulting LA measurements ranged from 20.2 to 45.4 α/khr/cm^2 (2.2X) although the replicate measurements for a single center were in very good agreement with less than 5% variation. This wide range of inter-center variation coupled with the excellent repeatability within a center is strong evidence of systematic error in the measurement. For the ULA samples, 11 of 23 measurements were within 1 standard deviation of the consensus mean and 7 were at or below

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background. However, the high level of counting uncertainty for ULA materials, with σ_{COUNT} as high as 210% of the consensus mean measurement, is sufficient to mask a systematic error as high as observed in the LA data. (The reader is referred to [8] or [7] for a more complete explanation of methods and terminology related to alpha counting.)

A survey of the methods used and subsequent discussions by the authors identified potential sources to explain the observed variations. The differences in the low energy discriminator setting for each counter, was thought to be the most likely hypothesis. This paper reports the results of a second round of data collection that was designed to assess this hypothesis.

II. METHODS

In order to examine the impact of the low energy discrimination setting, the second round (R2) of data collection utilized 2 samples. One sample had a broad range of alpha particle energies and the other was a monoenergetic source. If the discriminator setting was an important portion of the variation observed during R1 then the results from the first sample, similar to the energy range of the sample used in R1, should duplicate the variability of findings from that round and the results from the second should be immune to that variation as no events would be lost below the discriminator setting. Further, although it was not believed that differences in laboratory procedures had a large impact on the R1 results the authors decided to attempt to standardize the counting procedure to eliminate it as a possible source of variability.

A photograph of sample 1 is shown in Fig. 1 and its energy spectrum is shown in Fig. 2. The sample is a synthetic cordierite (approximately $Mg_2Al_4Si_5O_{18}$) based glass ceramic chosen for its appropriate level of emissivity and as a contrast to the metal samples used in R1. For alpha counters using

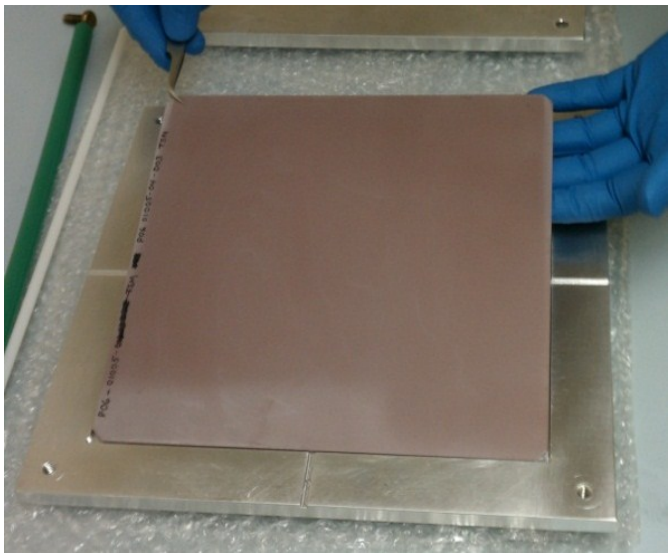

Fig. 1. Photo of the "thick" sample 1 (pink) being removed from its inner aluminum shipping container. The sample and shipping container were overwrapped with a foil bag purged with dry N_2 gas. The sample measures 20 cm on each side (400 cm^2) and has an emissivity of approximately 20 α/khr/cm^2.

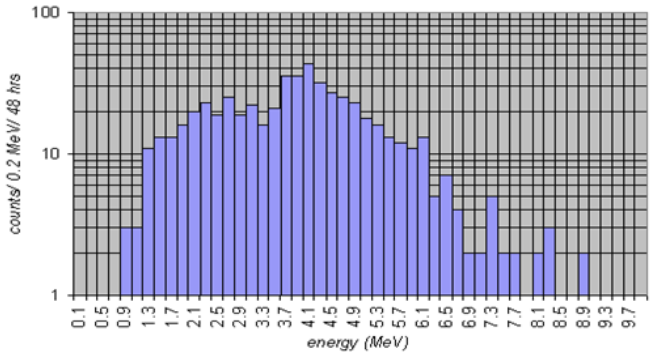

Fig. 2. Sample 1 spectrum demonstrating the smearing of emission lines to lower energies that is typical of a thick source. (Data courtesy M. Gordon.)

pulse analysis techniques the introduction of water vapor into the counting gas volume may increase electron mobility, resulting in a loss of counts [9]. Since the ceramic sample can readily adsorb water vapor and may later release it into the counting gas, it was dried before use and carefully managed during shipping. The sample was stored in a cleaned aluminum shipping container for mechanical protection (shown in Fig. 1) and overwrapped in a foil lined bag, purged with dry N_2 whenever it was opened and resealed. The ceramic sample was stored in this bag whenever it was not being counted. A permanent humidity indicator card was packaged inside the foil bag to ensure that the protocols were effective. For further protection the outermost shipping container was also sealed and provided with a desiccant package. None of the participants reported an indicator card showing excessive humidity during the study.

The spectrum in Fig. 2, recorded on an XIA ionization mode counter, is typical of materials with emissions from the bulk of the sample at depths comparable to the range of an alpha particle in that material, approximately 10 to 100 µm. Alpha decay results in discrete values of kinetic energy for the emitted particle but some of this energy is lost as the particle travels through the material. For samples that are "thick" compared to the alpha range the resulting spectrum at the surface is shifted by a variable amount towards lower energies. Most alpha counting samples of electronics materials are considered thick. Conversely, if the sample's alpha activity is confined to the surface or top few µm it is considered to be a "thin" source and will exhibit discrete spectral lines. Although a spectrum measured from a high activity sample may show some discernible structure that could be useful for isotope identification, the results from most LA and all ULA materials provide no useful identifying information for practical counting periods. The spectrum in Fig. 2 shows a general structure of a thick source but the detail needed to identify specific isotopes and their relative concentrations is not available. Due to the low chemical concentration and lack of spectral information, it is generally the case that the particular isotopes that are the source of emission for LA and ULA materials are not known.

The second sample is shown in Fig. 3. The active portion of the sample is a thin, electrodeposited layer of ^{230}Th on the

surface of the stainless steel substrate. The substrate is bonded to a ULA silicon wafer to simplify handling. A NIST traceable certificate of calibration was provided by the vendor [10]. The measured surface emission rate (2π geometry) is 94.0 α/min with a relative expanded uncertainty (k=2) of 2.8 α/min. (Note that this calibrated value is an emission rate with units of α/time rather than an emissivity in units of α/time/area. Unless noted otherwise, all references to sample 1 use the units of emissivity in α/khr/cm^2 and references to sample 2 use the emission rate in α/min.) Although the resulting count rate is much higher than would be recorded from an LA sample it is still well within the capabilities of all of the counters used in this study.

Considered as an emission rate, ceramic sample 1 registers approximately 0.1 α/min which is almost 3 orders of magnitude below that of sample 2. This emission rate is fairly typical for LA samples submitted for testing. Fortunately, the emission rate of sample 2 is low enough to be useful in these counters while still being high enough for characterization by NIST traceable equipment. The sample's energy spectrum is shown in Fig. 4 and demonstrates the expected discrete nature of a thin sample. The alpha particles emitted have E >3.5 MeV which is above the highest discriminator setting reported from R1.

Confidential data submission was also required by the participants for R2. The R1 protocol utilized an independent 3rd party and multiple confidential disclosure agreements to protect the participant's identities. A somewhat different procedure was used for R2 to assure confidentiality while reducing the administrative burden and costs. All data were collected and held by participants until the conclusion of counting. When all counting was completed each participant submitted the data as a set of plain text files along with the efficiency factor for each measurement. The files were electronically mailed to the study coordinator (J. Wilkinson) from a single, shared e-mail account to prevent identification. Prior to submission each participant self-assigned a 10 digit random number as a personal identifier and included it with the data. The identifier was subsequently used so that a participant may identify their own data in the published results. For publication clarity the random ID values have been replaced with single letters.

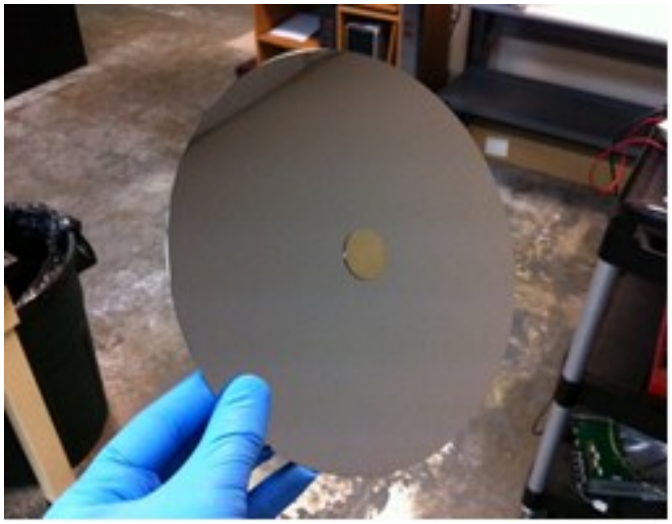

Fig. 3 Photograph of "thin" sample 2 (gold colored disk) mounted on a 200 mm ULA silicon wafer.

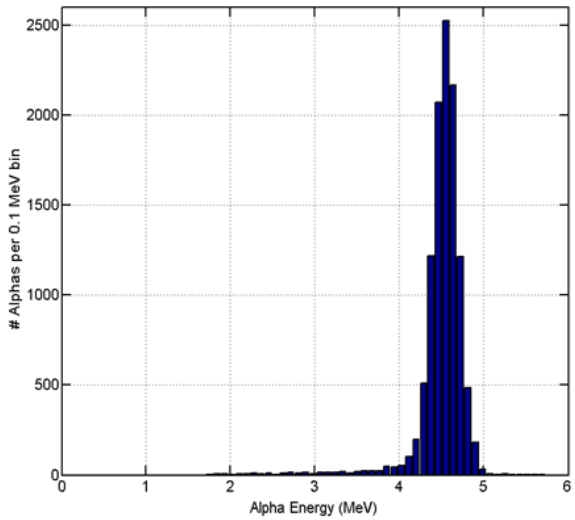

Fig. 4. The spectrum of sample 2 has all measured emissions above 3.5 MeV. Certified measurement of source emission rate is 94 +/-2.7 cpm. (XIA counter output, data courtesy B. McNally.)

Samples were sequentially mailed to each participant for counting. All counting was conducted using a written, standardized procedure sent with the samples that specified handling, counting times and reporting. The counting portion of the procedure is very briefly summarized here.

Sample 1 was counted for 48 hours and reported as 48 hourly values. Sample 2 was counted for 8 hours and reported as 48 values for every 10-minute period. For counters that required background subtraction a 48 hour background measurement was made prior to sample 1 and reported as 48 hourly values. Participants provided uncensored data for analysis to the study coordinator for a standardized analysis.

The data analysis removed readings at the start of the measurement period, as is customarily required, to allow the count rate to stabilize properly once the sample had been loaded. Rather than remove points for a specific period that may not be appropriate for all counters, the counting trends were tracked until the count stabilized. Specifically, data points were examined from the beginning and compared to the mean and standard deviation of the last 20 readings. Data at the beginning that were more than 2σ above the mean were discarded. Once a point had been found that was within 2σ of the mean that point and all subsequent points were retained for analysis. A mean of 1.9 data points were removed.

III. Results

Nine participants made measurements and one dataset was lost resulting in eight complete datasets. One of the remaining participants realized that a serious procedural error had been made in the measurement, recounted the samples and

TABLE 1
SUMMARY STATISTICS FOR THE ROUND 2 SAMPLES

Sample	Min	Max	Mean (μ)	S.D. (σ)	COV	Max/Min (σ/μ)
R1 LA	20.2	45.5	30.8	9.3	30%	2.3
R2 #1	13.7	29.9	21.8	5.9	27%	2.2
R2 #2	63.2	106	88.0	14.6	17%	1.7

Units for Min, Max and Mean values are α/khr/cm^2 for R1 and R2 sample #1 and α/min for R2 sample #2. The other values are dimensionless.

submitted a replacement dataset. The resulting 8 datasets are reported.

The thick sample results are illustrated in Fig. 5 and summarized in table 1 along with the summary statistics for the LA measurements from R1. The column "COV" is the coefficient of variation defined as σ/μ. Counting uncertainty is approximately 5% of the measured value. Considerable variation is evident and the results are not markedly different from R1 to R2, as expected. *Therefore*, t*he change in sample type had no apparent impact on the results nor did the standardized counting procedure*.

Results for the thin sample measurement are illustrated in Fig. 6 and summarized in the last row of table 1. The variation in count values, while somewhat smaller than the thick sample data and the LA results from R1, is still marked. Counting uncertainty has been reduced to less than 1% by the source's higher emission rate. Although there has been some reduction in the range of reported values it is clear that the role of the low energy discrimination threshold is limited. More troubling is the apparent measurement of count rate above the certified emission rate provided for this source. Alpha counters are inherently lossy instruments. Noise events that might be registered as sample counts are either characterized and removed during the background count phase or actively discriminated with pulse shape analysis. A variety of mechanisms have been described (e.g, see [7]) that lead to a loss of true counts. It is very unlikely that a significant increase in counts should be registered, particularly for this sample.

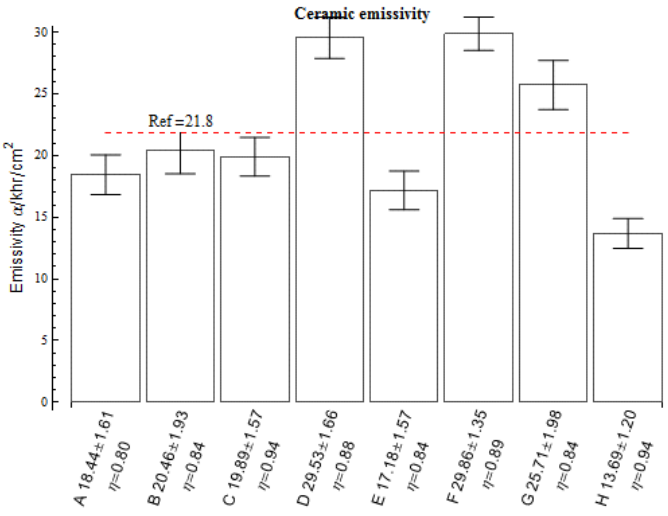

Fig. 5. Counting results for the ceramic sample (#1). The reference value is the consensus mean of the measurements. Error bars indicate the 1σ range due to counting uncertainty and do not include any estimate of systematic error. The mean count, 1σ count uncertainty and the counting efficiency (η) are reported below each bar.

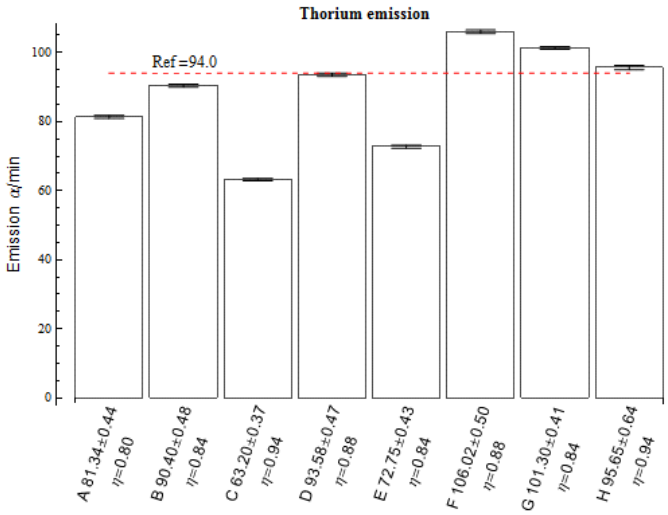

Fig. **6**. Counting results for the Th-230 (#2) sample. The analysis used the participant's reported counter efficiency value although this may not have been appropriate for this style sample (see text). The reference level is the source's certified emission rate.

IV. DISCUSSION

Lacking some independent measurement of the alpha particle emissivity for the ceramic sample it is not clear if the consensus mean measurement, one of the extreme value measurements or some other value entirely is closest to the true sample emissivity. If measurement disagreement stems from a random process then the consensus mean would be the most likely choice. On the other hand, a loss mechanism such as the low energy discrimination setting that introduces a proportional loss of counts that varies between participants would argue that the highest value is most likely closest to correct. And, of course, a combination of error mechanisms – or a lack of knowledge regarding the dominant mechanisms – leaves us to guess at the proper conclusion.

The alpha particle measurements from sample 2 permit the comparison to an independent standard and clearly demonstrate that the various measurements are all dominated by losses of various magnitudes. In this case, the best estimate of the true emissivity is the largest value. Although the sample types are somewhat different it is most probable to conclude that the measurements from sample 1 are similarly affected and that the true value is best represented by the largest value, which is 30 α/khr/cm^2 as measured by both D and F. The measurement uncertainty is then best quantified by comparing the ratio of the minimum and maximum values, rather than comparing with the mean value.

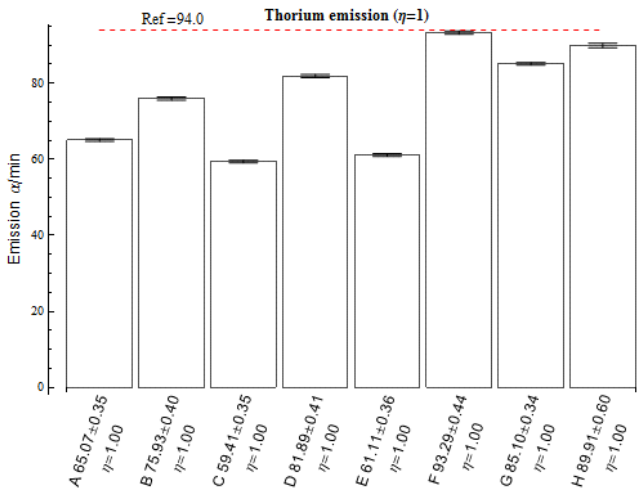

Fig. 7. Counting results for sample 2 with the efficiency (η) set to 1.0 to remove its effect from the analysis (see text).

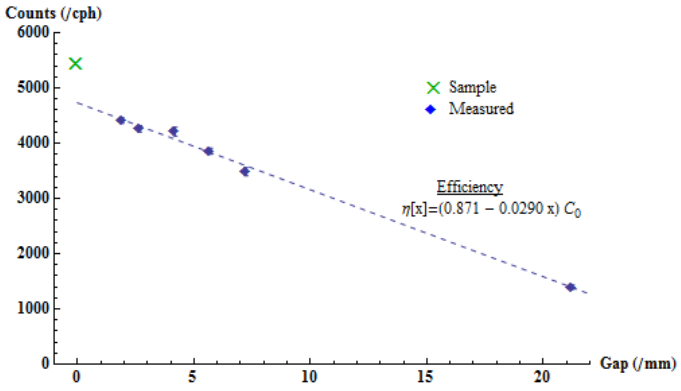

Fig. 9. Measurement of thin thorium sample's count rate as a function of the gap between the source surface and counter's entrance window. The measurement was not made with the source in direct contact with the window to avoid material transfer that would result contamination of the counter entrance window.

After further reflection on the results from sample 2 it was realized that the counter efficiency (η) reported by each participant might not be appropriate for that particular sample. A counter's efficiency is determined during a calibration process that measures the fraction of real alpha particle events that are emitted by a source but not registered by the counter for any of a variety of reasons. The monoenergetic character of sample 2 not only removes the effect of the low energy discrimination setting but it also reduces some trajectory related losses by concentrating the activity in a very small area near the center of the counter. Although the correct value for the efficiency of the counters is not known for sample 2 it is most likely that it should be higher than the value used for large area, thick samples most typical of alpha counting. The data for sample 2 were reanalyzed by setting η=1 and are shown in Fig. 7. For sample 2 in a windowless counter, used by 3 of the participants, this is likely to be the correct value as no events would be lost either to the periphery or due to energy discrimination.

The most notable effect of this adjustment is that all measurements are now at or below the calibrated value, with its uncertainty included, as expected for a set of counting measurements for a monoenergetic sample. The mean value is 76.5 α/khr/cm^2 and the COV is still 17% (as expected since it is not affected by renormalization of the data).

One loss mechanism for windowed counters, illustrated in Fig. 8, is affected by the gap between the emitting surface of the sample and counter's entrance window. The alpha counter can only register events that enter the counting gas volume above the entrance window with enough residual energy to create a signal that can be distinguished from noise. Alpha particles emitted at shallow angles may either fail to impact the entrance window or be absorbed within it. It is also possible for low angle alpha particles to lose all of their energy in the gas volume below the window after traversing approximately 4 or 5 cm laterally. For all of these cases, an increase in the gap dimension leads to increased losses of particles, particularly those emitted at shallow angles. To gauge the magnitude of this effect a series of measurements were made by M. Gordon using a windowed counter and a ^{230}Th source very similar to sample 2. Count data were collected with various separations between the entrance window and sample ranging from 2 to 21 mm. The results of the measurements are presented in Fig. 9. Error bars showing counting uncertainty are included on the plot but are generally smaller than the marker dimensions. The linear regression of the data, expressed as an efficiency, clearly shows that the efficiency at a gap of 0 is 87% and declines at a rate of 2.9%/mm.

The standardized protocol for this study did not specifically provide instructions for managing the gap nor did it ask for the value to be reported. Once the potential for the gap's impact was appreciated a confidential survey of the participants was made, asking for an estimate of the gap when the measurement was made. Seven of the 8 participants responded with results summarized in table 2.

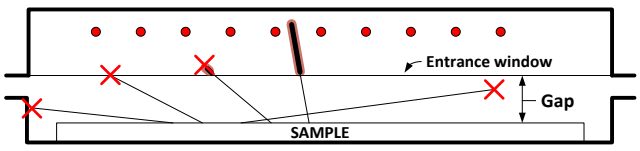

Fig. **8**. Loss of efficiency in windowed counter. Particles far from normal incidence may strike the side wall of the chamber, be absorbed in the entrance window or range out before entering the counting volume above the window. A large gap between the sample and window increases the fraction lost to these mechanisms, reducing counting efficiency.

TABLE 2
REPORTED VALUES FOR GAP AND THE ESTIMATED EFFICIENCY

ID	Gap (mm)	Estimated efficiency η
A	2.5	0.83
B	?	?
C	N.W.	1.0
D	2.3	0.84
E	6	0.72
F	N.W.	1.0
G	1.0	0.88
H	N.W.	1.0

Reported values for the measurement gap and the estimated efficiency using the model described in the text. Non-windowed (N.W.) counters are assumed to have an efficiency of 1.0 for this configuration.

Given the *post hoc* nature of this portion of the data collection along with the limited potential application of the gap model derived from measurements on a single counter it is not possible to responsibly draw quantitative results on this point. Suffice it to say that the variation seems to be reduced with application of this model to the windowed counters although data point C is still far removed from the remainder of the measurements.

Other loss mechanisms have been observed by various authors. For example:

- A contaminated sample tray will register counts during background determination. The contamination that is covered by the sample during the counting period will no longer be part of the background, leading to a reduction in the apparent emissivity. For ULA samples this has been observed to lead to apparently negative values for emissivity.
- Water vapor in the counting volume – potentially from room air, contaminated counting gas or sample outgassing – increases electron mobility and would reduce emissivity values in counters using active background discrimination. Fig. 10 illustrates this effect.
- A malfunctioning counter may have zones of inefficiency that are not apparent without careful mapping. The top portion of Fig. 11 presents a portion of the counting efficiency map of a counter that was discovered to be malfunctioning during detailed calibration testing while the bottom portion is the same data from a normally functioning counter. To emphasize the area that differs only the rearmost 4 cm of the counting area are shown. The top counter's efficiency for its rearmost 1 cm is less than 30%, most likely due to a malfunctioning anode wire.

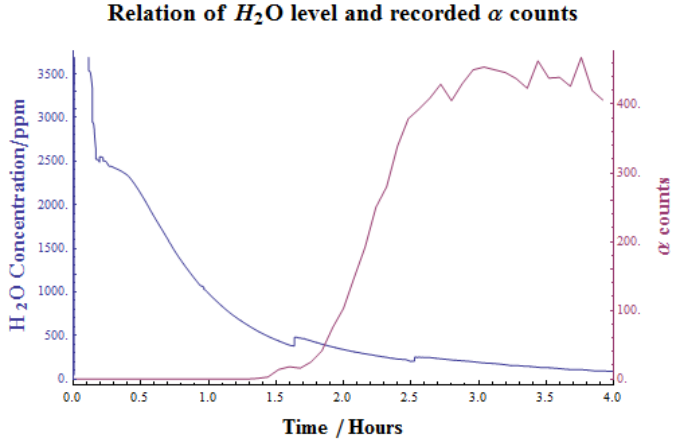

Fig. 10. Effect of water vapor on the alpha count. Moisture present in the counting volume may suppress the alpha count when utilizing active background discrimination.

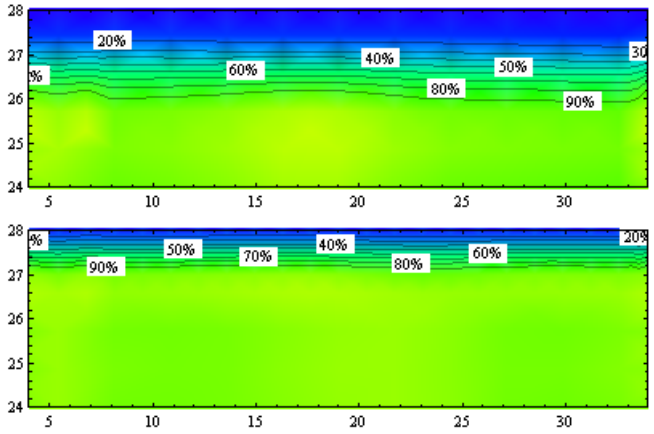

Fig. 11. Counting efficiency maps along the rear edges of a malfunctioning (top) and normal (bottom) counter.

The preceding list of loss mechanisms is certainly not exhaustive but serves to illustrates the variety of errors that have been observed. Processes to control these loss mechanisms, coupled with an appropriately determined counting efficiency are needed for accurate measurements. The authors recommend caution when attempting to make absolute measurements or to inter-compare measurements made by different laboratories.

V. CONCLUSIONS

This follow-up study has again demonstrated that the systematic errors in alpha particle emissivity measurements of LA materials contribute more to measurement uncertainty than the commonly cited uncertainty due to counting statistics. The major finding is a 1.7X variation (Max/Min) across the study participants for the ^{230}Th sample. Counting statistics and discriminator settings can be ruled out as a cause for this variation. A *post hoc* examination of the role of the sample to entrance window gap demonstrates that it is an important source of variation that was not controlled during the experiment. Some general conclusions may be drawn based on these findings and those of R1:

- The measurement of LA materials is generally repeatable within a center.
- Various loss mechanisms result in an underestimate of an LA sample's true emissivity by up to a factor of approximately 2.
- The use of alpha emissivity measurements for monitoring of manufacturing materials is likely to be effective for

determining trends and performance relative to previous samples at the same lab.
- When estimating alpha soft error rates the emissivity measurements for LA materials may be as little as 50% of the true value, thereby leading to an underestimate of the rate.
- All of the above conclusions are likely to be true for ULA materials although the higher degree of counting uncertainty and potential for other systematic errors may tend to degrade things further.

The lack of an appropriate calibration standard for these counters, along with an appropriate procedure for its use is a critical shortcoming for alpha emissivity metrology. Development and validation of a large area standard with suitable emissivity would be an important step forward and the authors encourage further research in this area.

VI. REFERENCES

[1] J. L. Autran, P. Roche, S. Sauze, G. Gasiot, D. Munteanu, P. Loaiza*, et al.*, "Altitude and Underground Real-Time SER Characterization of CMOS 65 nm SRAM," *Nuclear Science, IEEE Transactions on,* vol. 56, pp. 2258-2266, 2009.

[2] F. Wrobel, F. Saigne, M. Gedion, J. Gasiot, and R. D. Schrimpf, "Radioactive Nuclei Induced Soft Errors at Ground Level," *Nuclear Science, IEEE Transactions on,* vol. 56, pp. 3437-3441, 2009.

[3] M. S. Gordon, K. P. Rodbell, D. F. Heidel, C. E. Murray, H. H. K. Tang, B. Dwyer-McNally*, et al.*, "Alpha-Particle Emission Energy Spectra From Materials Used for Solder Bumps," *Nuclear Science, IEEE Transactions on,* vol. 57, pp. 3251-3256, 2010.

[4] B. M. Clark, "The Distribution and Transport of Alpha Activity in Tin," *Journal of Microelectronics and Electronics Packaging,* vol. 10, p. 10, 2013.

[5] R. C. Baumann and D. Radaelli, "Determination of Geometry and Absorption Effects and Their Impact on the Accuracy of Alpha Particle Soft Error Rate Extrapolations," *Nuclear Science, IEEE Transactions on,* vol. 54, pp. 2141-2148, 2007.

[6] H. Kobayashi, N. Kawamoto, J. Kase, and K. Shiraish, "Alpha particle and neutron-induced soft error rates and scaling trends in SRAM," in *Reliability Physics Symposium, 2009 IEEE International*, 2009, pp. 206-211.

[7] G. F. Knoll, *Radiation Detection and Measurement*, 4th edition ed.: John Wiley & Sons, Inc., 2010.

[8] J. D. Wilkinson, B. M. Clark, R. Wong, C. Slayman, B. Carroll, M. Gordon*, et al.*, "Multicenter comparison of alpha particle measurements and methods typical of semiconductor processing," in *Reliability Physics Symposium (IRPS), 2011 IEEE International*, 2011, pp. 5B.3.1-5B.3.10.

[9] A. Andronic, S. Biagi, P. Braun-Munzinger, C. Garabatos, and G. Tsiledakis, "Drift velocity and gain in argon-and xenon-based mixtures," *Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment,* vol. 523, pp. 302-308, 2004.

[10] Eckert and Ziegler Isotope Products, Inc., Valencia, CA.