LARGE AREA ALPHA SOURCES WITH A LIP: INTEGRAL COUNTING AND SPECTRAL DISTORTIONS

Lynne King, Ryan Fitzgerald* and Ronald E. Tosh
Physical Measurement Laboratory, National Institute of Standards and Technology,
Gaithersburg, MD 20899-8642, USA

Abstract

The detection efficiency for large area alpha sources with adjustable heights of a raised lip around the edge were measured by 2π gas-filled proportional counter. The variations in low-energy spectral shape were modeled using a Geant4 radiation and charge transport Monte Carlo simulation, to enable extrapolation of the spectrum to zero energy. COMSOL Multiphysics finite-element analysis was used to explore changes in the spectrum gain in the presence of a lip. It qualitatively reproduced an increase in peak height due to an increasing height of the source lip. A spectrum analysis procedure was developed to perform integral counting on sources with a lip. The experimental results were used to validate the model, which was then used to predict the changes in 2π counting efficiency for other source-lip geometries.

Keywords: proportional counter; large area sources; source geometry; Monte Carlo

Highlights

- Analysis method for proportional counter data for alpha sources with a lip.
- Monte Carlo analysis reproduces low-energy scattering shape and efficiency.
- Electric field calculations explain peak shift to higher energy.

*Corresponding author: ryan.fitzgerald@nist.gov

1. Introduction

Calibration of large area sources for surface emission of alpha and beta particles is essential for measuring performance of deployed instruments used in radiological protection, such as those used to monitor contamination resulting from nuclear accidents or release of radioactive materials (ISO, 1998; Unterweger and De Felice, 2015). Predominantly, such calibration sources are flat and either circular or rectangular, with radii or sides of a few centimeters and thicknesses of a few millimeters. The National Institute of Standard and Technology (NIST) large area proportional counters that are used for calibrations were themselves characterized, and are periodically validated, using sources of similar, flat geometry, making these detectors appropriate for calibrating flat sources. However, recently sources that have been sent to NIST for calibration sometimes take the form of a circular disc having a raised lip around the edge of several millimeters in height, simulating a cupped planchet used for smear counters (c.f. EAB-PL from Eckert and Ziegler, Berlin Germany)[1]. Measurements of 2π alpha emission rates of these sources produced energy spectra (Figure 1) that differed significantly from spectra of flat sources, in that an additional count peak appeared in the low-energy portion of the spectrum (Figure 1, region A), which is usually a local minimum. It was assumed that the additional counts were alphas emitted in the upward 2π hemisphere that then scattered off the lip, so that including the new peak in the integral counting would recover most or all of the 2π count rate. The present work is intended to discern any quantitative effect on

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[1] Certain commercial equipment, instruments, or materials are identified in this paper to foster understanding. Such identification does not imply recommendation by the National Institute of Standards and Technology, nor does it imply that the materials or equipment identified are necessarily the best available for the purpose.

the emission-rate measurements for these sources, by designing and measuring an alpha source geometry with a removable lip of adjustable height.

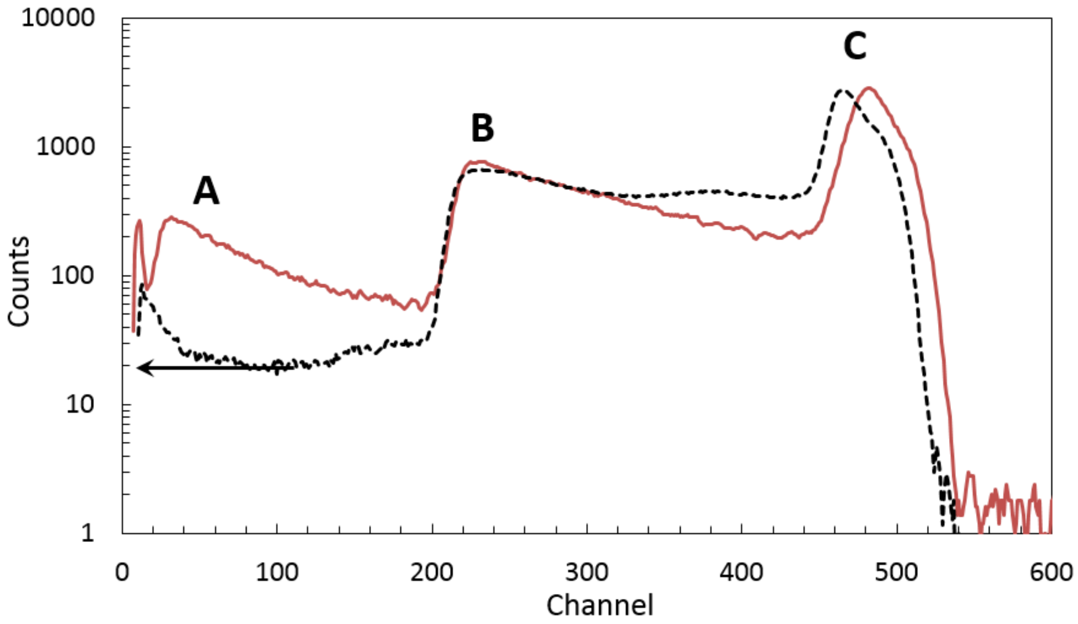

Figure 1 Spectra for an Am-241 planchet source with a lip (solid red line) compared to a flat Pu-239 source (dotted black line). For the source with a lip, Region A represents alpha particles that hit the lip. Shoulder B represents alpha particles that are emitted vertically and thus hit the top of the detector. Peak C is the full-energy peak, which is shifted in gain due to changes in the electric field caused by the lip. The black arrow indicates the usual horizontal extrapolation of the alpha tail to channel 0 for integral counting. See Section 2.1 and Figure 2 for details of the detector.

The analysis method normally used at NIST for surface emission rate measurements relies on horizontal extrapolation of the spectrum beginning at the spectral count minimum (Figure 1, region A), above the low-energy noise. However, that procedure would miss significant portions of the scattered spectrum for sources with a lip. Thus, a new procedure is required.

To develop and study a new procedure, a flat cylindrical Pu-239 source was used, to which rings could be added around the edge to produce lips of various heights. A new extrapolation method that would work for any ring height was devised and tested. Radiation transport modeling was validated by experiment and then used to predict detector response for other geometries.

During the study, it was found that adding lips to the source produced a secondary effect of increasing the spectrum gain (see high-energy peak in Figure 1). Although this gain increase did not have a significant effect on the integral counting, it could be important for spectroscopic studies. Therefore, the effect of the lip height of the electron multiplication of the detector was simulated using finite element analysis of the electric field in the detector for various source geometries.

2. Equipment and Methods

2.1 Experimental

The detector, electronics, and basic analysis method have been described by Hutchinson and Bright (1987) and Hutchinson (2004). In short, the 2π multi-wire proportional counter was run with flowing P-10 gas (standard 90 % argon / 10 % methane mixture by volume), at a nominal pressure of 0.1 MPa. The detector contains 19 wires (33 μm diameter, 30 cm length, 9.7 mm separation), which are electrically connected to form a single anode with voltage set at V_0= 750 V. The source and all walls of the detector form the cathode at ground potential. The anode signal is processed by a preamplifier and shaping amplifier and fed into a multichannel analyzer to record pulse-height spectra.

The radioactive source used for this work was a commercial wide area Pu-239 reference source, Figure 2. The nominal 2π alpha emission rate from the top surface of the source was 51 s^{-1}. The source is contained in an anodized aluminum foil of diameter 36 mm, embedded in the top surface of an aluminum disc of diameter 47 mm. The thickness of the anodized aluminum layer is about 5 μm.

To create sources with a lip, a set of aluminum rings were made having 8 different heights, *h*, between 0.6 mm and 9.5 mm. These rings were mounted one at a time, or else stacked, on the disc source. The inner diameter of the rings was chosen to leave a small gap of width, *w*, up to 4 mm, between the active area of the source and the ring. Figure 2 shows a schematic of the detector containing the source with a ring.

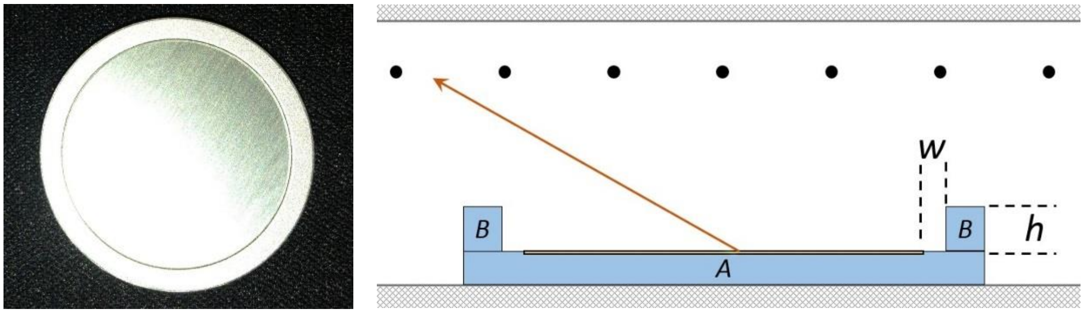

Figure 2 (Left) Photograph of Pu-239 source. (Right) Zoomed-in side-view of the 2π proportional counter, shown to scale except for the wire diameter, which is actually 33 µm. A lip is created by placing a ring, B, of height, h, (4 mm as shown) on top of the Pu-239 disk source, A. There is a radial gap of width, w, between the active area of the source (yellow inset in A) and the inner surface of the lip. All surfaces except the wires are at ground potential. The wires are at potential V_0. The entire detector contains 19 wires, but only the central 7 are shown. An example Pu-239 alpha track is illustrated by the orange arrow.

Data were acquired for the same source with and without the rings. Typically, 5 repeat measurements of 1 000 seconds each were recorded. Additional longer runs lasting 20 000 seconds were taken for the source without rings. Background spectra were also recorded. Spectra were corrected for background before performing further analysis. First the typical integral-spectrum-counting analysis method was performed (Hutchinson 2004), which involves placing a low-energy cut on the spectrum above the detector noise floor and then extrapolating the low-energy tailing of the alpha peak to channel zero using a linear function of slope 0 (horizontal) or else one that slopes down to the origin. In this work, a horizontal extrapolation was used, with the count level set by averaging over about 20 channels in the region of local count minimum below the alpha shoulder

(See Figure 1). Then an alternative method was developed to extrapolate the low-energy tail using an exponential function. Each spectrum was analyzed separately and the results for a given geometry were then averaged.

The alternative method of extrapolation was devised specifically for the spectra that these lipped sources generate. Here it was assumed that all the counts in region A (Figure 1) were from alpha particles emitted in the forward 2π hemisphere that hit the lip, still depositing some energy in the P-10 gas. The hypothesis was that an exponential extrapolation of the tail of this scattering peak from around channel 20 downward to channel 0 would recover the true 2π emission rate, as measured by the usual procedure with no lip. An exponential function was fit to the tail of the low-energy peak. This equation was used to predict how many counts would be in the channels from the line back to channel zero, eliminating low-energy noise. The area in question was small relative to the total spectrum, e.g. about 0.5 % for h = 0.

2.2 Modeling

To better understand the spectral distortions caused by sources with a lip, and to be able to predict the effect on integral counting results, computer models were constructed.

Preliminary calculations indicated that the increase in counts in the low-energy spectrum were due to alpha particles hitting the lip, which was then simulated with a Geant4 radiation transport model (Agostinelli et al, 2003). The model was made using

Geant4.9.5, with the standard electromagnetic physics package. Particle production cuts were set to 10 μm and the step limit was 10 μm in the detector gas and 0.2 μm in the source. The geometry was set based on measurements of the detector and technical drawings of the source and rings. The general shape of the Pu-239 depth distribution in the source was taken from Berger et al. (1996), with the parameters tuned to the specifications of this source and to match the tailing of the spectra. The simulation was run for $1 \cdot 10^6$ decays of Pu-239 for each experimental geometry, as well as for other geometries not measured experimentally. The Monte Carlo random number generator was constructed to produce the same set of initial Pu-239 atom positions in the source and initial alpha decay trajectories for each h. That way, small differences in the spectra among geometries were revealed. The simulated spectra were analyzed in the same way as the experimental spectra.

The increase in gain of the experimental alpha peaks with increasing h appeared to be due to changes in the electric field in the counter, due to the lips. The gain is not critical for integral counting, but could be important for spectroscopic methods. The gain was hypothesized to be due to increased electron multiplication caused by changes in the electric field in the presence of a lip. The multiplication was calculated following the general method of Diethorn (1965), (see also Charpak, 1968; Knoll, 2000), but instead of assuming the electric field of a cylindrical geometry, the electric field was simulated using a COMSOL Multiphysics (COMSOL, Inc., Burlington, MA, USA) finite element analysis model. The model assumes that electron multiplication occurs only in the region of space near each wire, where the electric field exceeds a critical value, E_c. Each free electron moving through that region will undergo a multiplication event after traveling across a voltage drop of D, which depends on the nature of the counting gas. For P-10 gas, we assumed a voltage drop of D = 23.6 V, and a critical electric field of E_c= 4.80 · 10^4 V/cm (Knoll, 2000). The COMSOL model was solved to find the critical potential, V_c, at the radius from a wire corresponding to E_c. The multiplication M was then calculated as

$$M = 2^{(V_c - V_0)/D}. \tag{1}$$

The values of M for a given wire were averaged over the central 50 mm length of wire, where most of the electrons produced by the slowing down of the alphas were collected, according to the Geant4 model. Variation of M with wire position was estimated by modeling the effect on a wire centered over the planchet and another wire centered over the lip (i.e. along a tangent to the annulus formed by the lip).

3. Results

Experimental and Geant4-simulated spectra are shown in Figure 3. The bottom panel shows both the original extrapolation method (horizontal from the local minimum, which is around channel 60 for no-lip data) and the new method (exponential from the lower edge), which were applied to this data to determine the best way to account for the change in spectral shape (both methods are described in section 2.1). When the standard horizontal method was used, many of the counts in the low energy region were cut out, significantly reducing the estimated emission rate, which clearly demonstrates this method is not appropriate for these sources. Using the exponential trend-line for each background-subtracted-measurement resulted in values that are much closer to the nominal emission rates measured without a lip (Table 1). On average, the measured 2π alpha emission rate for sources with a lip was 0.5 % higher than for the same source without a lip, with a standard deviation of 0.3 % among lip heights. There was no significant trend in extrapolated emission rate value vs h.

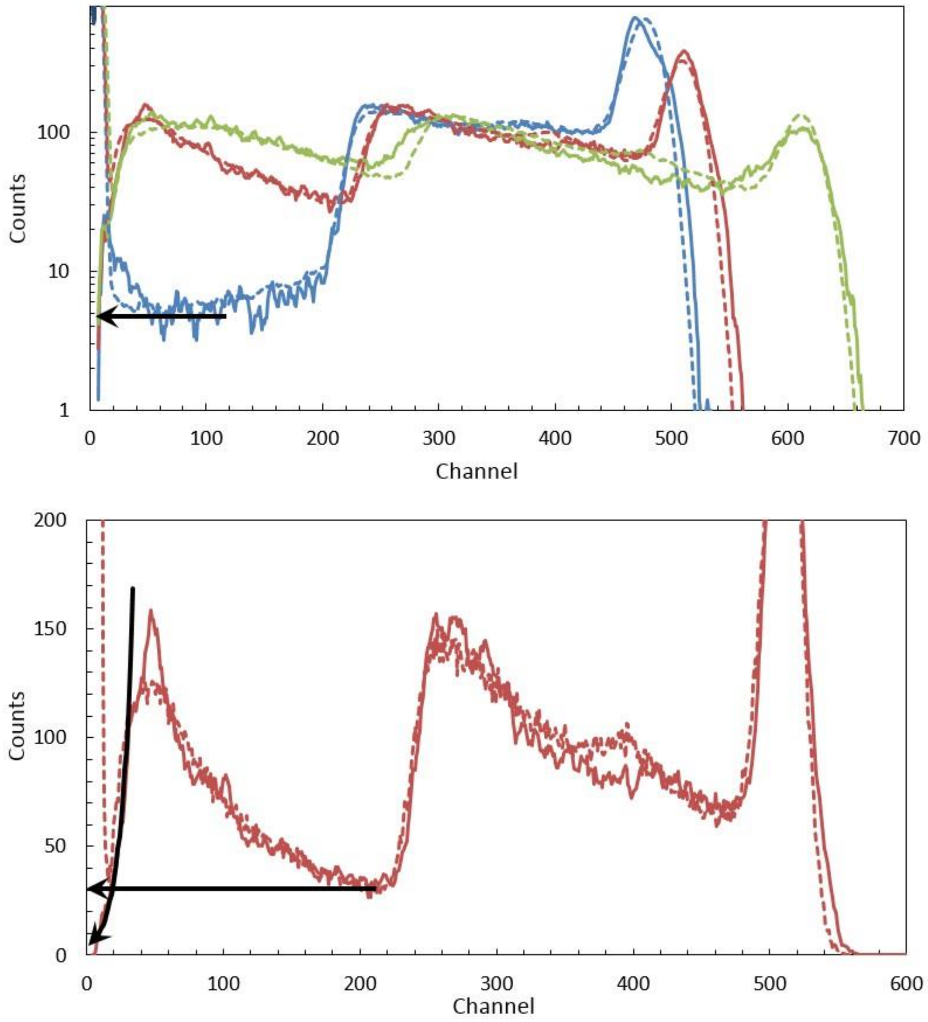

Figure 3 Top: Experimental (solid lines) and Geant4-simulated (dashed lines) spectra from left to right of lip heights h = 0 (no lip, blue), h = 4.8 mm (red), and h = 9.5 mm (green). The pulse-height gains of the Geant4 spectra were chosen to match experimental peak positions. Horizontal extrapolation shown for h=0 (arrow). Bottom: Illustration of extrapolation for standard horizontal extrapolation and new exponential extrapolation (arrow) for h = 4.8 mm data.

Table 1 Experimental and simulated relative changes in the derived 2π emission rate, Δε, by traditional horizontal extrapolation method and new exponential method. The standard deviation for 5 measurements is given in parentheses. The random sampling uncertainty for the simulation (sim) data is about 0.05 %.

	horizontal	exponential	
h **(mm)**	**$\Delta\varepsilon$ (%)**	**$\Delta\varepsilon$ (%)**	**$\Delta\varepsilon_{sim}$ (%)**
0.6	-0.5	0.3 (4)	0.5
0.8	-1.3	0.5 (4)	0.5
1.3	-2.8	0.4 (2)	0.5
2.5	-5.5	0.3 (3)	0.3
3.0	-6.0	1.1 (4)	0.5
4.0	-7.3	0.5 (1)	0.7
4.8	-14.2	0.3 (4)	0.8
9.5	-12.2	0.8 (3)	0.8

The Geant4-simulated spectra generally match the experimental data, and additional simulations for alpha particles emitted at fixed angle, θ, from the normal vector to the source surface add valuable qualitative spectral information, as follows. Referring to Figure 3 (top), the full-energy peak corresponds to alpha particles emitted at $47° < \theta < 90°$, whereas the shoulder around channel 240 corresponds to $\theta = 0°$ at which alphas reach the top wall of the detector with significant kinetic energy remaining. In the presence of a lip, alpha particles with $\theta \rightarrow 90°$ produce the low-energy peak in the spectra around channel 50. Below channel 50, there is an upturn in counts heading toward channel 0.

The Geant4 model was further used to predict efficiencies for additional geometries. Spectra simulated by fixing h at 3.2 mm, while varying the width, w, between the radius of the active area of the source and the inside of the lip from w = 0 mm to w= 4.0 mm are shown in Figure 4. Surprisingly, for $w = 0$, the bias in 2π alpha emission rate measurement is only $\Delta\varepsilon$ = 0.04 %, demonstrating that good measurements are still possible. The biggest bias was for $w = 1.0$ mm, for which the exponential extrapolation would over-estimate the emission rate by 0.41 %, apparently because the extrapolation region is strongly affected by the low-energy noise.

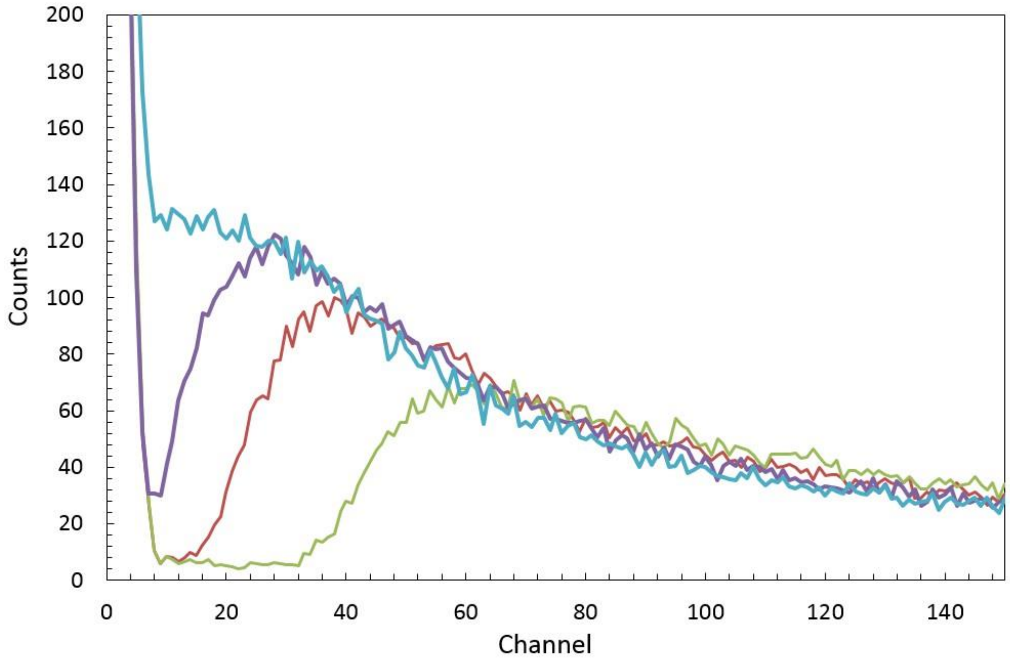

Figure 4 Geant4 simulations for sources with $h = 3.2$ *mm and varying widths between active radius and inner radius of lip. From left-to-right are spectra for w of 0 mm (blue), 1.0 mm (purple), 2.5 mm (red), and 4.0 mm (green). Exponential extrapolations yield biases of -0.02 %, 0.24 %, 0.41 % and 0.04 %, respectively.*

The electric field simulations predicted an increase in gain in the presence of a lip, due to an increase in the critical radius for multiplication around each wire, which increases $V_c - V_0$ in the exponent of Equation 1. The magnitude of M depends on how it is sampled radially around each wire, though the change in M vs. h is less sensitive to this. The simulated relative change in M vs. h is shown in Figure 5, along with the experimental changes in alpha peak gain, ΔP, expressed in %. Qualitatively, both gains increase quadratically with h. However, ΔM calculated with COMSOL is significantly smaller than experimental ΔP. A quantitative model prediction of ΔP would likely require further integrating the COMSOL and Geant4 simulations such that individual electron and ion trajectories could be followed and the full response of the pulse-shaping electronics could be simulated.

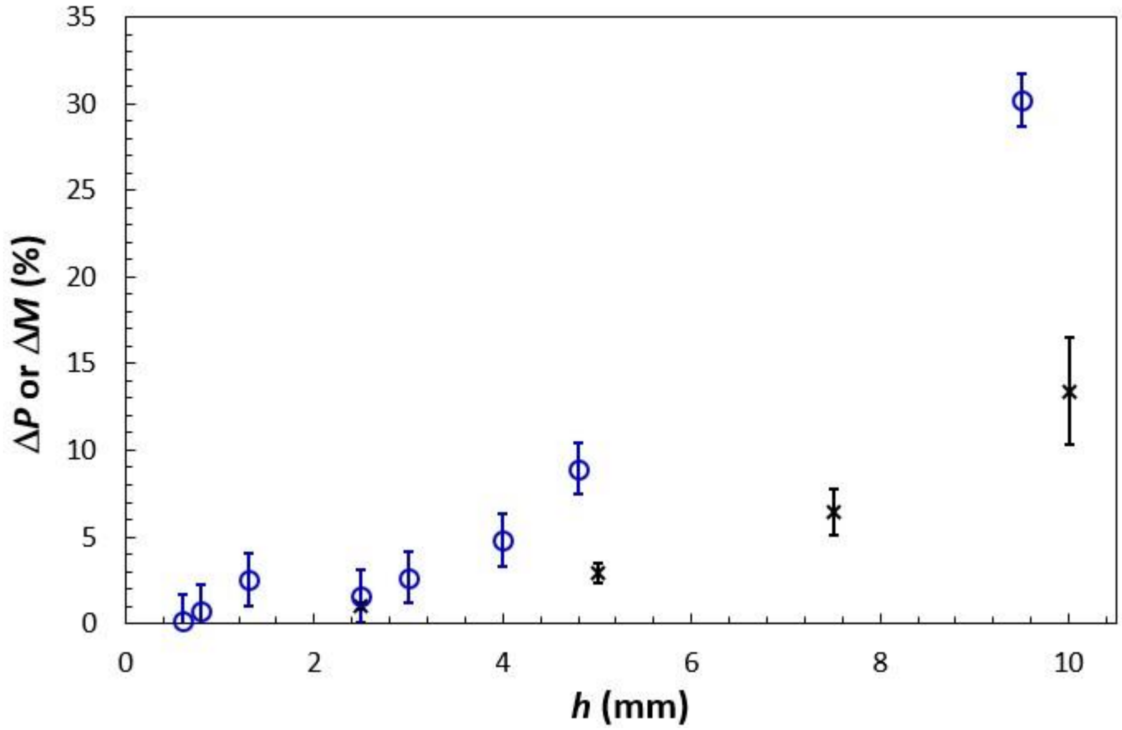

Figure 5 Percent gain shift of the alpha peak, ΔP, (blue circles) as a function of lip height, h. Uncertainty bars represent the standard deviation of repeated measurements. Black line is a quadratic least-squares fit. The black crosses are COMSOL model calculations of the change in gain, ΔM, with bars representing the variation of calculated values for different wires in the multiwire proportional counter.

4. Conclusions

It was determined that alpha-sources with a lip have a different spectral shape and that current methods of extrapolation exclude a significant number of these counts. A new method for customizing extrapolations using trend-lines was developed and tested. The new method is superior to the historically used method for these sources and should be considered for calibrations of alpha-sources with a lip. Without further modification, the new method is expected to produce about a 0.5 % bias in the result, due to convolution of low-energy noise with the true counts from alphas that deposit only a small fraction of their energy in the P-10 gas before hitting a lip. If the low-energy behavior of the spectrum for a certain type of source is well-known, then spectra could be corrected for this before or after extrapolation, reducing the bias.

The Geant4 model reproduced the major characteristics of the experimental spectra and the changes in those characteristics as h was varied. The shortcoming of the model is in the low-energy spectrum in which the simulation only accounts for alpha straggling, conversion electrons, Auger electrons, x-rays, and gamma-rays. However, the upturn toward $E \rightarrow 0$ may be related to other processes, such as recoiling atoms from Rutherford scattering, which will require additional model development to simulate.

Another use of the model would be to deduce the active radius of a large-area source by putting a ring around the source and comparing the shoulder of the spectra with those simulated (e.g. Figure 3) to extract w. Imaging techniques can be used to find w for flat sources. The presence of the lip distorts the image making it less useful in determining the precise value.

Finally, it is worth noting that the measurand considered in this work is the 2π alpha emission rate directly above the surface of the source. However, in the presence of a lip, the emission rate of alphas that escape the entire source (passing above h) will be lower than the emission rate at the surface. Therefore, if the source is used to calibrate a detector with a window (rather than sitting inside a 2π counter), a correction would be necessary to obtain the effective emission rate at the detector face. The correction could be approximated by subtracting the area in region A from the integrated spectrum. For $h = 4.8$ mm and $w = 2.0$ mm, the correction would be -22 % or -21 % from the experimental or Geant4 spectra, respectively. Alternatively, by modifying the Geant4 model to record the fraction of alphas that stop in the lip, a correction of -27 % is derived.

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