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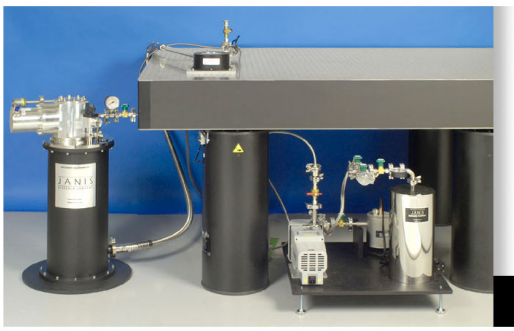

REVIEW OF SCIENTIFIC INSTRUMENTS **88**, 035108 (2017)

A method for calibrating the relative gamma-ray light yield of plastic scintillators

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Currently we are investigating the inclusion of organotin compounds in new polystyrene scintillator materials to improve full gamma-ray energy sensitivity. Accurate calibration of the relative light yield from the newly developed scintillators is crucial to assess merits of compounds and chemical processes used in the scintillators' development. The full energy gamma-ray peak in a measured gamma-ray spectrum is commonly used in calibrating the relative light yield. However, the Compton continuum in the newly developed plastic scintillators is measured with much better efficiency and statistics and is found to be the best spectral feature that can be exploited for expeditious calibration of the relative light yield. In this study, we present a spectral gain matching of measured and simulated spectra, using a spectrum rebinning technique, to determine the Compton edge in a measured Compton continuum for accurate relative light yield calibration. The Compton edges determined using this technique were found to be within 1.2% of their theoretical estimates. *Published by AIP Publishing.* [http://dx.doi.org/10.1063/1.4978288]

I. INTRODUCTION

The inclusion of heavy metals in common plastic scintillator materials has been a topic of investigation for enhancing full energy gamma-ray sensitivity.[1–4] We have been investigating organotin loaded polystyrene scintillators for potential gamma-ray spectroscopy applications. Material and performance aspects of the scintillators have been reported in our earlier publication.[5] Research and development of these plastic scintillators require accurate calibration of light yield from a gamma-ray interaction to assess merits of compounds and chemical processes used in scintillator development and to assess the scintillation efficiencies of the newly produced scintillator samples. Typical light yield calibration or measurement is accomplished by analyzing fully absorbed energy or the photo peak of the incident gamma-ray. However, the Compton continuum is measured with better statistics and efficiency than the photo peak in the newly developed plastic scintillators. It was believed that the Compton edge could be exploited for expeditious and accurate light yield calibration. The use of the Compton edge also enables the direct comparison of the relative light yields of organotin loaded scintillators with pure polystyrene based scintillators. For an ideal radiation detector with an excellent energy resolution, the Compton edge is a sharp edge representing a discrete energy. This, however, is not the case with plastic scintillators due to their energy resolution. The Compton edge is smeared and requires analysis to determine the position of the Compton edge in the measured gamma-ray spectrum.

In the present work, we demonstrate a method for calibration of plastic scintillators' relative light yield using the Compton edge. Spectral gain matching between measured and simulated gamma-ray spectra was implemented for the calibration. Monte Carlo N Particle, version 5, (MCNP5) code was used to simulate gamma-ray spectra in the newly developed plastic scintillators. We start by giving a brief overview of past efforts in using the Compton edge for energy or pulse height calibrations. Following, we will discuss the technique implemented in this work. Finally, we will present results from the technique implementation.

II. COMPTON EDGE FOR ENERGY AND LIGHT YIELD CALIBRATION

A. Brief review of past efforts

A number of previous efforts on Compton edge localization in a gamma-ray spectrum can be found in a literature search. We selected several interesting works and review them in this report. The earliest work of interest was by Pringle *et al.*,[6] which followed the discovery of sodium iodide scintillator (NaI(Tl)) by Hofstadter.[7] Pringle *et al.* associated the Compton edge, which they termed as the upper energy limit of the Compton distribution, with the inflection or the Compton maxima in the NaI (Tl) pulse height calibration. It was noted that the full energy photo peak was not resolved using NaI(Tl). This was mainly due to stated limitations associated with light collection and photomultiplier tube (PMT) variance. The technique implemented by Pringle *et al.* was first introduced by Siegbahn[8] to analyze data collected using a beta spectrograph. However, the use of the inflection point or the Compton maximum can make accurate calibration of the relative light yield very challenging. Chikkur and Umakantha[9]

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implemented a Gaussian curve fitting approach to determine the Compton edge measured using liquid scintillators with varying solute concentrations. Accordingly, the high energy side of the Compton continuum was Gaussian fit with varying Gaussian parameters for each solute concentration as shown in Fig. 1. The centroid, $\bar{a}$, and standard deviation, σ, from each fit were then used to determine the Compton edge channel as $a_c = \bar{a} + 1.177\sigma$. Although the theory behind the use of the expression 1.177σ was not explained, it represents the half width at half maximum (HWHM) of the Gaussian fit. It can be deduced from Fig. 1 that it would be problematic to get a fairly reproducible segment in the spectra for a Gaussian fitting and therefore reproducible $\bar{a}$ and σ. It can also be inferred from the works of Deitze *et al.*, described below, that the expression for a_c with the HWHM is not an accurate representation of the Compton edge in a measured pulse height spectrum. Besides questions on the accuracy of the equation used, it is likely that the approach will be difficult to implement in measurements that involve multiple scattering events.

Dietze[10] and later Dietze and Klein,[11] unlike their predecessors, compared Monte Carlo simulation with experimental data to determine the Compton edge position in measured pulse height spectrum using NE-213 liquid scintillator. Accordingly, Dietze *et al.*, based on Monte Carlo simulation, characterized the energy or channel separations between locations of the Compton maximum, the Compton edge, and the Compton half maximum. The characterization was carried out using three varying parameters: the energy resolution, the detector dimension, and the incident gamma energy. Tables and curves were then generated to help in locating the Compton edge in a measured data. The Compton maximum and half maximum from a measured pulse height spectrum were extracted and then used to determine the energy resolution. Subsequently the Compton edge was found using tables and curves. Dietze *et al.* demonstrated the accuracy of their technique using coincidence measurements. The technique sounds reasonably accurate for a known detector dimension, gamma energy, and energy resolution. Dietze *et al.* did not explain how the extraction of the Compton maximum and half maximum

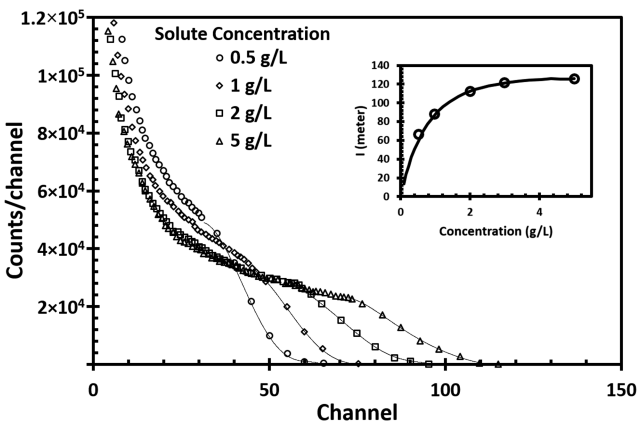

FIG. 1. Measured pulse height distribution using liquid scintillators with varying solute concentration, Reproduced with permission from G. C. Chikkur and N. Umakantha, Nucl. Instrum. Methods Phys. Res. 107, 201 (1973). Copyright 1973 Elsevier. Solid curves represent a Gaussian fit to the high energy side of the Compton continuum. The inset shows the Compton edge pulse height as a function of solute concentration.

was done in the measured pulse height spectrum. However, Gaussian fitting of the high energy side for the extraction of these parameters, as was done by Chikkur *et al.*, might be problematic in detectors with significant multiple scatterings.

Several works addressing the use of coincidence techniques for Compton edge localization have been demonstrated in the past.[11–15] It is understood that coincidence techniques can be the best strategy to accurately locate the Compton edge. This approach may decrease the detection efficiency and commonly involves the use of a secondary detector and electronic modules. Consequently, the use of a coincidence setup for routine energy calibration or spectroscopy may be unnecessary.

Hohara *et al.*[16] also used Monte Carlo simulation with experimental data to determine the Compton edge. The simulation was made using the Klein-Nishina formula[17] and electron stopping power within a plastic scintillator. Chi-squared minimization was used to fit the experimental data with simulated data. The resolution of the simulated data was varied to get the lowest chi-squared value and therefore the best fit to the experimental data. Thinner samples ranging from 2 to 10 mm were used in their simulation to avoid multiple scatterings. The fit near the Compton maximum was good. A discrepancy between the fit and the experimental data was observed at the lower energy region likely due to electron escape unaccounted for in their simulation.

Siciliano *et al.*,[18] based on the interest for homeland security applications, used what they termed as the ratio algorithm to determine the Compton edge in Polyvinyl Toluene (PVT) detected spectra. Accordingly, a ratio of the counts at the Compton maximum and at the Compton edge is determined using MCNP5[19] simulated spectra for gamma-ray energies of interest in a PVT detector. A simple detector setup was used in their simulation and the simulated energy spectra (ESs) were smeared using MCNPs Gaussian Energy Broadening (GEB) card that approximates the resolution of the simulated detector. The ratio determined using the simulation was used in the measured data to determine the Compton edge by multiplying the ratio with the Compton maximum in the measured spectrum. Siciliano *et al.* noted in their work that there is a slight dependence of their approach implemented with the energy resolution of the detector. Kudomi,[20] with an interest in the rare beta (β) and $\beta\beta$ decay measurement applications, similar to Hohara *et al.*,[16] used an analytical expression based on the Klein-Nishina formula[17] to fit Geometry And Tracking (GEANT3.2.1)[21] simulated gamma-ray spectra to determine the Compton edge in plastic scintillators. Kudomi noted a discrepancy between expected and evaluated Compton edges that worsened as the size of the plastic detector increased. Kudomi attributed the observed discrepancy to multiple scatterings that were not accounted by the fitting analytical function. Several more works related to calibration of low atomic number detectors using Compton edge localization may be found in Refs. 22–25.

B. Technique implemented in the present work

In the present work, we demonstrate the use of MCNP5 simulated gamma-ray pulse height spectra (SPHSs) to

determine the Compton edge in a plastic scintillator measured gamma-ray pulse height spectra (MPHSs). The MCNP5 simulation was specific to the measurement setup and included all elements in the setup that may impact the observed spectrum. The SPHSs were generated by smearing MCNP5 simulated energy spectra (ESs) with varying energy resolution. The novelty in the present work is the iterative gain matching approach based on root mean square error (RMSE) minimization. The gain matching is done by weighted iterative rebinning and comparison of the SPHS with the MPHS until a minimum RMSE is obtained. The ES is simultaneously rebinned with the SPHS until the minimum RMSE is achieved. The location of the Compton edge in the ES finally determines the Compton edge in the MPHS. It was found that the Compton edge determined is almost independent of the energy resolution used in the SPHS. Initially, the SPHS and the ES are in energy units while the MPHS is in channel units. After the gain matching, both the SPHS and ES are effectively converted into channel unit without the requirement for energy versus channel calibration curve or varying energy resolution for a best fit, as was done by Hohara *et al.*[16] This has an advantage in the development of new scintillators, as in our case, where light yield and energy resolution may significantly vary from one scintillator sample to another. It excludes the need for interpolation or separate simulation of each sample as was done by Dietze and Klein.[11] The approach can also help avoid complications that may arise due to multiple scatterings in larger volume detectors. Details on the effort and analysis carried-out are presented below.

1. Experimental setup and measurement

Light yield from newly developed plastic scintillator samples was measured using a Hamamatsu[26] R1828-01 PMT coupled to an Ortec[27,28] 575-A spectroscopic amplifier and 928-MCB multi-channel analyzer. Samples produced were polished for smooth coupling with the PMT. Each sample was wrapped with a Teflon tape for efficient light collection before coupling to the PMT using polydimethylsiloxane optical grease. The measured samples were approximately 0.5 cm thick and 2.54 cm in diameter. Cs-137, Mn-54, and Na-22 check sources with gamma energies 662, 834, and 1274 keV, respectively, were used in the measurements. Samples characterized and gamma-ray sources used both have right cylindrical geometries and were framed using plastic holders to establish the same concentric axis for reproducibility. This also allowed an easy reproduction of the experimental setup geometry in the MCNP5 simulation. Plastic frames used have insignificant impact in the measured spectra. However, they were accounted for in the MCNP5 simulation described below. MPHSs from each sample were then acquired for about 5 min, as that was sufficient for statistically meaningful and reproducible data analysis.

2. MCNP5 simulation

MCNP5 was used to simulate gamma-ray energy spectra using gamma-ray energies of interest. Compositions of plastic scintillators and gamma-ray scattering elements near to the measured plastic scintillator were included in the MCNP simulation. Coupled gamma-electron transport was made to generate the absorbed energy spectrum in the simulated sample. It was necessary to do the coupled gamma-electron transport to account for electron escape from 0.5 cm thick samples. The absorbed energy spectrum was further processed to generate the SPHS using the MCNP5 GEB card that allowed for the inclusion of energy resolution of the measured sample. Figure 2 shows SPHS with 12%, 18%, and 25% energy resolutions and ES at 662 keV (Cs-137) gamma-ray energy. A pure polystyrene-based material with no organotin loading was used in the simulation. As can be seen in Fig. 2, the material used is efficient enough to resolve the low energy X-ray peaks from the Cs-137 source used in the simulation. Features related to multiple scatterings within the simulated scintillator are also visible at the right side of the Compton edge in the ES. For subsequent light yield calibrations, it was necessary to account for all measurable spectral features in the simulated spectra that may be from nearby gamma-ray scattering elements or radio isotopes used in the measurement.

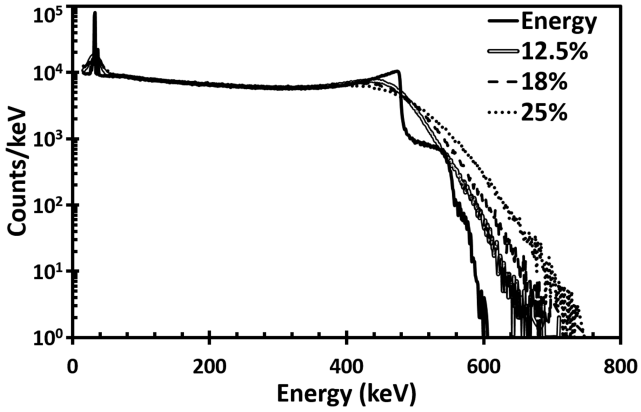

FIG. 2. MCNP5 simulated energy and pulse height spectra using Cs-137 source. Pure polystyrene-based material was used in the simulation. The simulated energy spectrum was smeared with resolution ranging from 12.5% to 25%.

3. Measured and simulated spectra gain matching

As is mentioned above, measured and simulated spectra gain matching is done by successive rebinning of SPHS and comparing it with MPHS until minimum RMSE is determined. Rebinning of the simulated spectra, SPHS and ES, may expand or shrink the total number of bins in the spectra. Total counts normalization of the measured and the simulated spectra were made before proceeding in gain matching. Rebinning of SPHS and ES, each with n number of bins, into m number of bins of the MPHS is carried out iteratively. Representing the original SPHS or ES by a vector Y and after rebinning the transformed vector by X, the rebinning process is described using Eq. (1),

$$X_{i,k} = W_{i,j} * Y_{j,k}, \quad i = 0, 1, \ldots, m-1,\ j = 0, 1, \ldots, n-1, \text{ and } k = 1, \tag{1}$$

where W is the weighting factor that assigns fraction of the original bin that goes into the destination bin. W is a two

dimensional matrix and is given by Eqs. (2a)–(2c),

$$W_{i,j} = 1, \quad if\ j \le i * \left(\frac{MB}{LB}\right), \tag{2a}$$

$$W_{i,j} = i * \left(\frac{MB}{LB}\right) - (j-1), \quad if\ j > i * \left(\frac{MB}{LB}\right), \tag{2b}$$

$$W_{i+1,j} = j - \left(i * \left(\frac{MB}{LB}\right)\right),\ i = i+1, \quad if\ j > i * \left(\frac{MB}{LB}\right),$$
$$j = 0, 1, \ldots, MB-1, \tag{2c}$$

where MB and LB stand for more bins and less bins, respectively. If the total number of bins in MPHS is greater than that in SPHS/ES, then MB will be equal to the total number of bins in the MPHS and LB will be equal to the total number of bins in SPHS/ES, else vice versa. $W_{i,j}$ is an $LB \times MB$ matrix calculated for index, j, ranging from 0 to MB-1. During the calculation, the index, i, is incremented in steps to a maximum value of LB, when the condition in (2b) is satisfied. For a case of a vector Y expansion in the gain matching, $W_{i,j}$ transpose $(W_{i,j}{}^T)$ is used in (1) with appropriate normalizations in successive rebinnings.

MPHSs are given in counts versus channel units while SPHS and ES are given in counts versus energy units. Conversion from energy to channel units or vice versa is not necessary before proceeding to rebinning. Spectra matching through rebinning of the simulated spectra effectively makes the conversion from energy unit to channel unit. Calculated RMSE shows either an increasing or decreasing trend. The successive rebinning of the simulated spectra is terminated after the RMSE passes a global minimum and starts increasing. Care must be taken to avoid a local minimum that may result in the termination of rebinning. At the global minimum, the location of the Compton edge in the rebinned ES determines the Compton edge in the MPHS. The Compton edge determined is in channel units and represents the pulse height or the light yield recorded from the plastic scintillators at the Compton energy.

4. Validation of the spectra gain matching approach

Validation of the above outlined technique was made by carrying out measurements using NaI(Tl) and Bismuth Germanate (BGO) scintillators. These inorganic scintillators give the advantage of producing fairly linear energy calibration curves above 100 keV gamma-ray energy using measured photo peaks. Therefore, the Compton edge determined in channel units using the above outlined technique can be associated with energy units. Similar to plastic scintillators, MCNP5 was used to generate the SPHS and ES for the inorganic scintillators that were used in the spectra gain matching and determine the Compton edges as described in Subsection III. Compton edges determined through the analysis were then compared with calculated theoretical Compton edges.

III. RESULTS AND DISCUSSION

A. Spectra gain matching

Figure 3 shows measured and simulated spectra before (a) and after (b) spectra gain matching for one of our newly produced plastic scintillator with no organotin loading. Gain matched spectra in Fig. 3(b) were determined at the global minimum of the RMSE that was calculated after each successive rebinning. SPHS smeared at 12.5% energy resolution was used in the gain matching. Figure 4 shows the trend in RMSE as a function of the number of bins for the spectra analyzed. Some differences between the measured and simulated spectra are evident after rebinning (Fig. 3(b)). These differences are attributed to differences in energy resolutions between MPHS and SPHS. The MPHS as shown has a slightly better energy

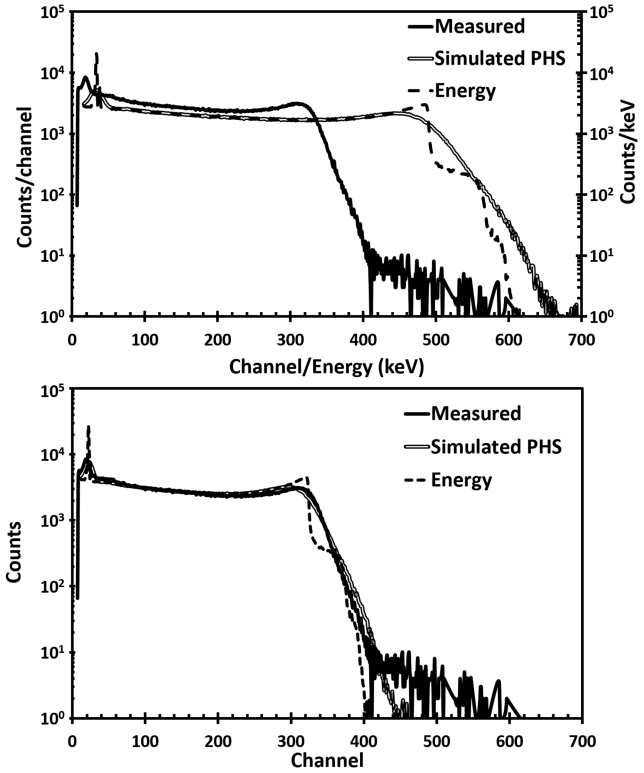

FIG. 3. (a) Measured and simulated spectra before gain matching for selected pure polystyrene scintillator with no organotin loading. Cs-137 source was used in both the simulation and the measurement. (b) Measured and simulated spectra after gain matching. Some differences are observed between the measured and the simulated PHS that are due to differences in energy resolution.

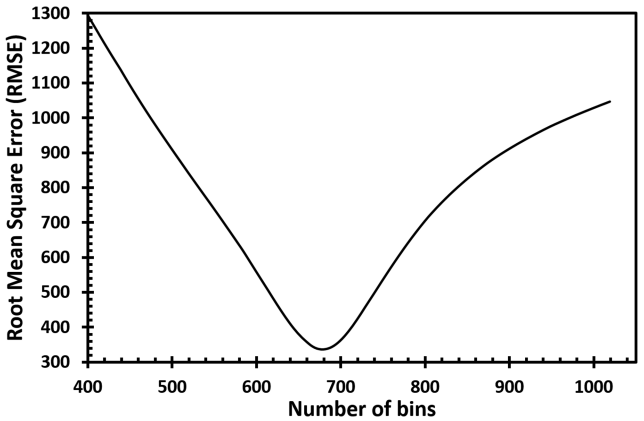

FIG. 4. Root mean square error (RMSE) as a function of number of bins in successively rebinned simulated spectra. Rebinned spectra were selected at the global minimum of the RMSE.

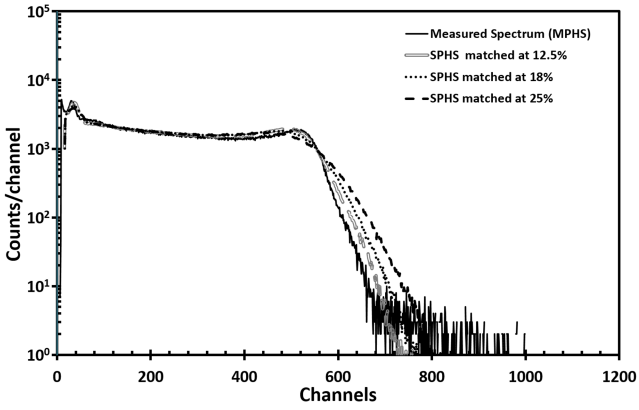

FIG. 5. Simulated spectra (SPHS) with energy resolution at 12.5%, 18%, and 25% gain matched with the measured spectrum (MPHS). Pure polystyrene-based material and Cs-137 source were used in both the simulation and the measurement.

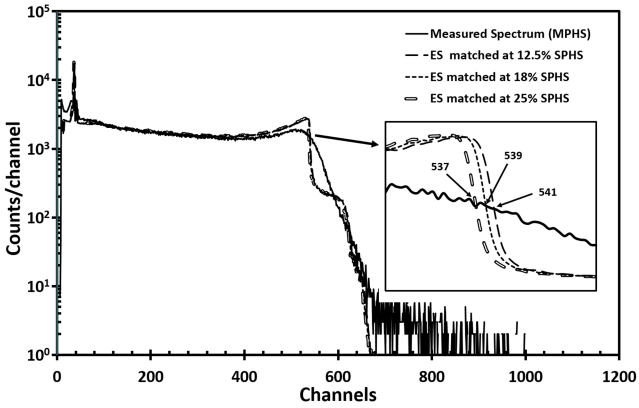

FIG. 6. Comparison of Energy Spectra (ES) matched to measured spectrum (MPHS) using simulated spectra (SPHSs) with varying energy resolution. Compton edge determined varies insignificantly as a function resolution used in SPHS as shown in the inset. Features related to multiple scattering events are evident at the right side of the Compton edge in the ES. Pure polystyrene-based material and Cs-137 source were used in both the simulation and the measurement.

resolution. This, however, brings the question if SPHS resolution has some impact on the final Compton edge determined thus on the relative light yield. To evaluate the extent of any impact, SPHSs at 12.5%, 18%, and 25% (Fig. 2) were used separately in the spectra gain matching as shown in Fig. 5. The corresponding ESs, and Compton edges, matched using the different resolution spectra are shown in Fig. 6. It can be seen in the inset with exploded view that there is an insignificant drift in the edge (<1%) as the energy resolution changes significantly. This confirms the negligible effect of the SPHS energy resolution on the present technique.

B. Technique reproducibility

Reproducibility of the implemented technique was tested using variable experimental amplifier gain settings. A scintillator sample with no organotin loading was measured at five different amplifier settings using a Cs-137 source. Amplifier gain settings at 3, 4, 5, 6, and 7 were used. Figure 7 shows the MPHS and the matched PHS and ES spectra for representative

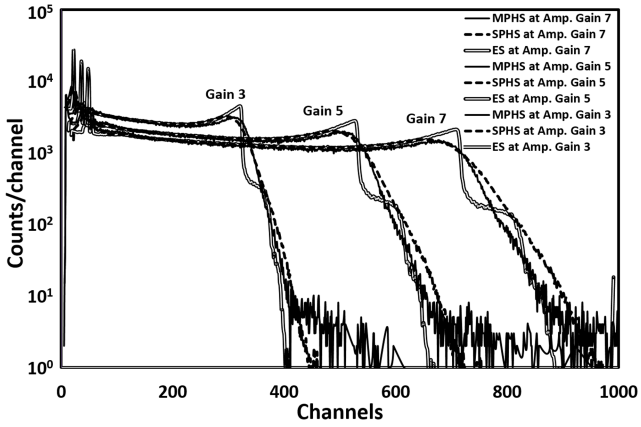

FIG. 7. Gain matched MPHS, SPHS, and ES at three experimental amplifier settings. Pure polystyrene-based material and Cs-137 source were used in both the simulation and the measurement.

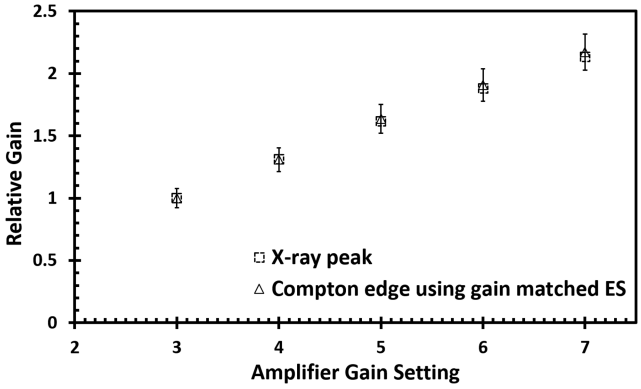

FIG. 8. Relative gain determined using Gaussian fit of MPHS X-ray peak and gain matched ES Compton edge. Amplifier settings were incrementally varied from 3 to 7 in the measurement.

amplifier gain settings at 3, 5, and 7. The relative gain in the measured spectrum at each amplifier setting was determined using the X-ray peak centroids evaluated using Gaussian fitting. Alternatively, the Compton edges determined from the gain matched ES were also used to determine the relative gain. Figure 8 shows relative gains as a function of amplifier settings. Good agreement was found between relative gains determined based on the X-ray peaks and gain matched ES Compton edges to within 2%.

C. Technique validation

Compton edges determined after implementing the technique outlined Secs. III A and III B are given in channel units. It is necessary to confirm that the evaluated edges agree with the expected theoretical values in energy units thus validating the technique. NaI (Tl) and BGO inorganic scintillators were used for validation of the implemented technique. Similar to the analysis made using plastic scintillators, these inorganic scintillators were simulated using MCNP5 at 662, 834, and 1274 keV gamma-ray energies. Both detectors were also used in the measurement and calibration at the same energies. Measurements were made using the same PMT and electronics used in the plastic scintillators measurements.

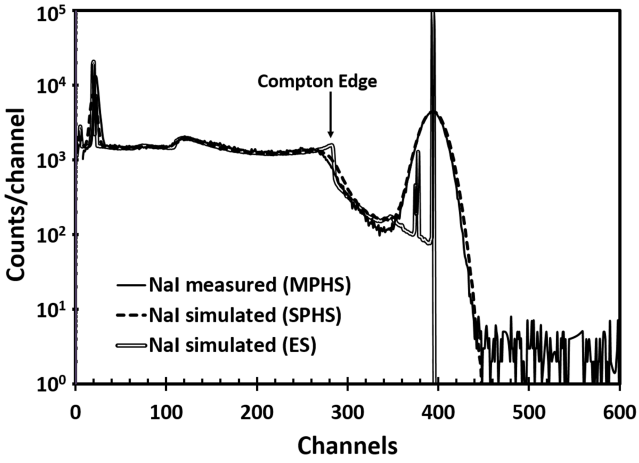

FIG. 9. NaI (TL) detector gain matched MPHS, SPHS, and ES using Cs-137 source. The Cs-137 (662 keV) photopeak and the escape peaks are clearly visible in the ES spectrum as distinct lines at the right side of the Compton edge.

Calibrations of the detectors were made using a linear fit to peak channels versus peak energy data. The peak channels were obtained by Gaussian fitting of the full energy peaks detected in the spectra. Spectra gain matching of simulated and measured spectra was made to determine the Compton edges in channel units. The linear fit from the calibration was then used to determine the measured Compton edges in energy unit. It is assumed in the analysis that the detectors used have reasonably proportional light yield in the energy range above 100 keV.[29,30]

Figures 9 and 10 demonstrate the spectra gain matching made at 662 keV for the NaI(Tl) and BGO detectors. For BGO simulated spectra, both SPHS and ES show some discrepancies in the Compton continuum counts compared to the measured data. These differences are likely due to differences between the simulation and the measurement scattering environments. Table I tabulates theoretical Compton edges together with

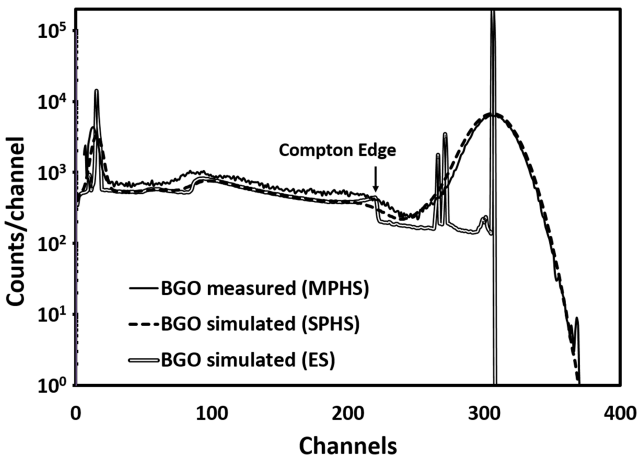

FIG. 10. BGO detector gain matched MPHS, SPHS, and ES using Cs-137 source. Discrepancies in the Compton continuum counts are evident between the simulated and the measured data, likely due to differences in the scattering environment. The Cs-137 (662 keV) photo peak and the escape peaks are clearly visible in the ES spectrum as distinct lines at the right side of the Compton edge.

TABLE I. Evaluated Compton edges using NaI (Tl) and BGO detectors.

	NaI (Tl)		BGO	
Source (keV)	Evaluated (keV)	Difference (%)	Evaluated (keV)	Difference (%)
Cs-137 (478)	473	1.05	480	0.31
Mn-54 (638)	632	0.94	641	0.44
Na-22 (1061)	1067	0.57	1048	1.23

those determined using the present technique. Good agreements were found that were within 1% for NaI (Tl) and 1.2% for BGO detector. Assuming a linear response for NaI (Tl) and BGO in the energy range of interest likely contributes to the slight discrepancies observed. Although a good gain match at the photo peaks in Figs. 9 and 10 is observed, a gain mismatch is evident between the simulated and measured spectra at the X-ray peaks that is mainly due to nonlinearity in the measured spectra at the X-ray energies. However, results are very encouraging and promise the use of the present technique in relative light yield calibration of low resolution detectors such as plastic and liquid scintillators.

IV. CONCLUSION

In the present work, we demonstrated a technique for the determination of a Compton edge, therefore the relative light yield, in a measured plastic scintillator gamma-ray spectrum. Spectral gain matching through rebinning of MCNP5 simulated pulse height and energy spectra with the measured spectrum was instrumental in the determination of the Compton edge. The technique implemented was found to have insignificant dependence on the energy resolution of the simulated spectra. Results obtained are encouraging and reasonably accurate in determining the Compton edge. Future studies will investigate the possibility of a nonlinear rebinning based on measured nonlinearity of the scintillators to further improve the accuracy of the implemented technique. We believe that the present implemented technique can be useful for relative light yield calibration in detectors with low energy resolution such as plastic and liquid scintillators.

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