SAND2015-10999J

Plastic Scintillators Light Yield Energy Calibration

Wondwosen Mengesha, Patrick L. Feng, Joseph G. Cordaro, Mitchell R. Anstey,
Nicholas R. Myllenbeck, and Daniel J. Throckmorton

***Abstract*–Currently we are investigating the inclusion of organotin compounds in polystyrene material to improve plastic scintillators full gamma-ray energy sensitivity with the ultimate goal of achieving spectroscopy. Accurate evaluation of light yield from the newly developed scintillators is crucial to assess merits of compounds and chemical process used in the scintillators development. Full gamma-ray energy peak in measured gamma-ray spectrum, resulting from total absorption of gamma-ray energy, would be ideal in evaluating the light yield from the new scintillators. However, full energy sensitivity achieved thus far is not statistically viable for fast and accurate light yield energy calibration from the new scintillators. The Compton edge in measured gamma-ray spectrum has been found as an alternate gamma-ray spectrum feature that can be exploited for characterizing the light yield energy from the newly developed plastic scintillators. In this study we present technique implemented for accurate light yield energy calibration using the Compton edge. Results obtained were very encouraging and promise the possibility of using the Compton edge for energy calibration in detectors with poor energy resolution such as plastic and liquid scintillators.**

***Index Terms*—Plastic scintillators, Compton edge, light yield, energy calibration, spectral matching, gain normalization, FFT, Savitzky-Golay.**

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 application. Research and development of these plastic scintillators require accurate characterization of light yield from a gamma-ray interaction with the scintillator samples produced. Typical light yield measurement is accomplished by analyzing fully absorbed energy or the photo peak of the incident gamma-ray. However, current plastic scintillators under development don't have sufficient sensitivity to resolve the fully absorbed energy with good counting statistics in a short measurement time. It may require few hours of counting to generate statistically significant counts from fully absorbed energy. Using the Compton continuum is an alternate approach for an accurate light yield evaluation in a short time of measurement. The Compton continuum has a characteristic edge that represents the maximum energy deposited when an incident gamma scatters at 180 degree. For an ideal radiation detector with an excellent energy resolution, the Compton edge is a sharp edge representing a discrete energy. The new scintillators under development have been measured to have energy resolutions ranging from 10 to 15% at 662 keV. Localizing the ideal Compton edge under such conditions is challenging.

Several works have been reported in the past to address similar challenges [5-6]. Siciliano et al [5], 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 spectrum. Accordingly a ratio of the maximum count nearby the Compton edge or what they referred as the Compton maxima and the counts at the Compton edge is determined using Monte Carlo N Particle version 5 (MCNP5) [7] code 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 were smeared using MCNPs Gaussian Energy Broadening (GEB) card that approximates the resolution of the simulated detector. Ratio determined using the simulation was used in the measured data to determine the Compton edge by multiplying the ratio with the Compton maxima in the measured spectrum. Siciliano et al. noted in their work that there is slight dependence of their approach implemented with the energy resolution of the detector. Kudomi [6], with the interest in the rare beta (β) and ββ decays measurement applications, used analytical expression based on Klein-Nishina formula [8] to fit Geometry And Tracking (GEANT) version 3.2.1 [9] code simulated gamma-ray spectrum to determine the Compton edge in plastic scintillators. Kuodomi noted discrepancy between expected and evaluated Compton edges that gets worse as the size of the plastic detector increases. Kuodomi attributed the observed discrepancy to multiple scatterings that were not accounted by the fitting analytical function.

In the present work we demonstrate the use of MCNP5 simulated gamma-ray spectrum to determine the Compton edge in plastic scintillators measured gamma-ray spectrum to allow light yield energy calibration. Spectra gain matching of simulated and measured pulse height spectra using root mean square error (RMSE) minimization was implemented. Following the spectra gain matching, Fast Fourier Transform (FFT) [10] analysis was made to determine the Compton edge. MCNP5 simulation in the present work was specific to the measurement setup and included elements in the setup that may impact the observed spectrum. Details on the effort and analysis carried-out are presented.

The authors are with Sandia National Laboratories, Livermore, CA 94550 USA (telephone: 925-294-6264, e-mail: wmenges@sandia.gov)

Sandia National Laboratories is a multi-program laboratory managed and operated by Sandia Corporation, a wholly owned subsidiary of Lockheed Martin Corporation, for the U.S. Department of Energy's National Nuclear Security Administration under contract DE-AC04-94AL85000.

This work was supported by Department of Homeland Security-Domestic Nuclear Detection Office (DHS-DNDO).

II. METHODS

Light yield from newly developed scintillator samples was measured using Hamamatsu [11] made R1828-01 photomultiplier tube coupled to Ortec [12] 575-A spectroscopic amplifier and 928-MCB multi-channel analyzer. The measured samples were approximately 0.5 cm thick and have 2.54 cm diameter. Cs-137, Mn-54, and Na-22 check sources with energy 662, 834, and 1274 keV respectively were used in the measurements. Light yield calibration of the samples was made using collected gamma-ray spectrum and MCNP5 simulated pulse height and energy spectra. Spectral gain matching and FFT were implemented to determine the light yield associated with the Compton edge.

A. MCNP5 Simulation

MCNP5 was used to simulate gamma-ray energy spectra using gamma-ray energies of interest. Composition of plastic scintillator 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 absorbed energy spectrum in the simulated sample. It was necessary to do the coupled gamma-electron transport to account electron escape from 0.5 cm thick samples. The absorbed energy spectrum was further processed to generate simulated gamma-ray pulse height spectrum using the MCNP5 GEB card that allows inclusion of energy resolution of the measured sample. Fig. 1 shows simulated 662 keV (Cs-137) gamma-ray energy and pulse height spectra at 12, 18, and 25% energy resolutions. As can be seen in Fig. 1, X-ray peaks are evident in the simulated plastic scintillator. For subsequent light yield calibration, it was necessary to account all measurable spectral features in the simulated spectra that may be from gamma-ray scattering elements nearby the measured samples or radio isotopes used in the measurement.

B. Spectra gain matching

Measured and simulated spectra gain matching was done by successive rebinning of simulated pulse height and energy spectra. Rebinning of the simulated spectra may increase or decrease the total number of bins in the simulated spectra.

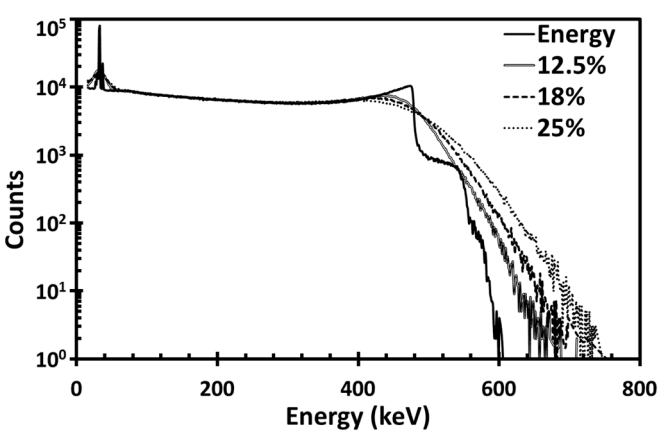

Fig. 1. MCNP5 simulated energy and pulse height spectra. Simulated energy spectrum was smeared with resolution ranging from 12.5 to 25%.

Measured spectra are given in counts versus channel units while simulated spectra 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. Total counts normalization between the measured and simulated spectra was made before the start of the successive rebinning. Counts in each bin are used as weighting when carrying out the rebinning. It is important to confirm that the successive rebinning of the simulated spectra do not alter the simulated gamma-ray spectral features. After each rebinning step, root mean square error (RMSE) between measured and rebinned simulated pulse height spectra is calculated. The RMSE shows either an increasing or decreasing trend. The successive rebinning of the simulated, both the pulse height and energy, 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. It is obvious that at the global minimum the Compton edge of the rebinned energy spectrum may be very close to the measured spectrum Compton edge. The Compton edge may subsequently be determined using one of the well-established spectrum unfolding techniques. FFT technique [10] was implemented in the present work and is briefly described below.

C. Fast Fourier Transform (FFT)

Deconvolution of the measured pulse height spectrum collected from the newly developed plastic scintillators was made to determine the Compton edge. FFT was implemented using gain matched simulated pulse height and energy spectra to determine the smearing function that is instrumental in unfolding the measured spectrum. Accordingly the simulated PHS may be given in terms of the energy spectrum, E, and smearing function, R, as:

$$PHS = E \otimes R \tag{1}$$

In frequency domain R may be determined using a simple algebraic division as:

$$R(f) = \frac{PHS(f)}{E(f)} \tag{2}$$

The FFT approach although fast and straight forward, is highly sensitive to noise and high frequency artifacts and may require iterative low pass filtering without impacting the low frequency spectral features. Prior to the implementation of the FFT measured and simulated spectra were treated with iterative Savitsky-Golay (SG) [13] low pass filter. During the FFT unfolding, Weiner filter [10] derived from power spectral densities (PSD) of measured spectra was used to further suppress noise and artifacts related features in the deconvolved spectrum. R determined using the simulated spectra can then be used to deconvolve the measured pulse height spectrum in similar form as in (2) by replacing the ratio with the measured spectra and R to determine the energy spectrum, E, therefore the Compton edge.

Validation of the above outlined technique was made using NaI(Tl) and BGO scintillators. The technique was implemented on measured spectra from these scintillators using different gamma-ray energies. Compton edges determined through the analysis were compared with the expected theoretical Compton edges.

III. Results and Discussion

A. Spectra gain matching and FFT unfolding

New scintillators developed and characterized commonly have different light yield and material composition and therefore varying energy resolutions. It was of interest to know if technique implemented in the present work, in particular the FFT, would be dependent on the energy resolution of the scintillators. MCNP5 simulated spectra shown in Fig. 1 were used to verify the FFT dependence, if any, on the energy resolution of the scintillators. Accordingly simulated PHS at 25% resolution was used to determine R in (2). R determined was then used on the spectra at 12.5 and 18% resolution to determine the corresponding energy spectra and therefore the Compton edges. Prior to application of the FFT, the spectra were treated with the SG filter. During the process of FFT implementation Weiner filter, derived from the power spectral density (PSD) of the pulse height spectra, was applied to further suppress impacts from noise and high frequency artifacts. Spectral gain matching was not needed as simulated spectra were used for the analysis. Results from the FFT implementation are shown in Fig. 2. Compton edges unfolded have the same energy as the simulated. Some differences are observed in the counts of the unfolded spectra near by the Compton edge. However, it is evident from the plot that the energy resolution of the measured scintillators has minimal impact in the Compton edge determined using FFT unfolding.

Fig. 3 shows measured and simulated spectra before (a) and after (b) spectra gain matching. Total counts normalization of the measured and the simulated spectra were made before proceeding in gain matching. Gain matched spectra in Fig. 3(b) were determined at the global minimum of the RMSE that was calculated after each successive rebinning. Fig. 4 shows the trend in RMSE as a function of number of bins for the spectra analyzed. Some differences between the measured and simulated spectra are evident after rebinning (Fig. 3(b)) that are mainly due to differences in the energy resolutions. These differences, however, have minimal impact in the final result as shown in Fig. 2. The simulated spectra shown in Fig. 3(a) are in energy units. Rebinning the spectra in Fig 3(b)

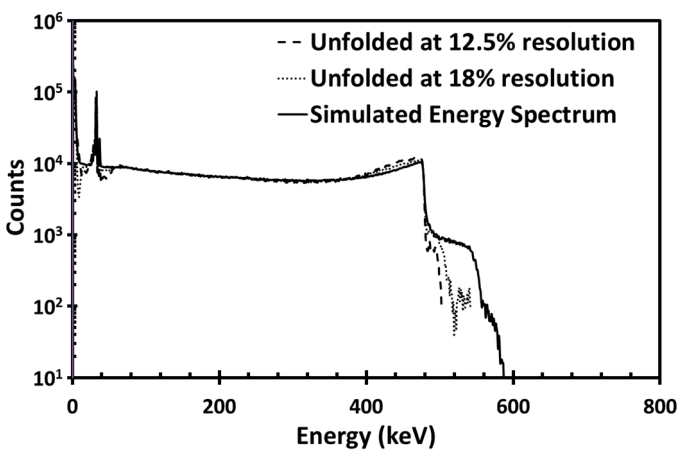

Fig. 2. FFT unfolded energy spectra. Unfolded Compton edges have the same energy as the simulated confirming no dependence of the FFT technique on energy resolution.

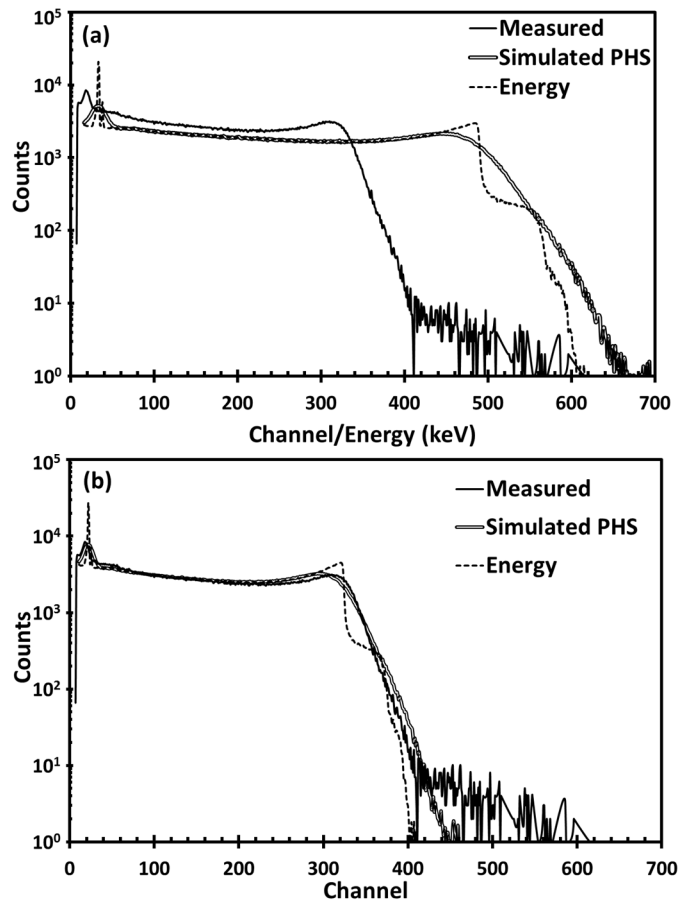

Fig. 3. (a) Measured and simulated spectra before gain matching. Both simulated spectra, pulse height and energy, are gain matched before proceeding to FFT unfolding. (b) Measured and simulated spectra after gain matching. Some differences are observed between the measured and simulated PHS that are due to differences in energy resolution.

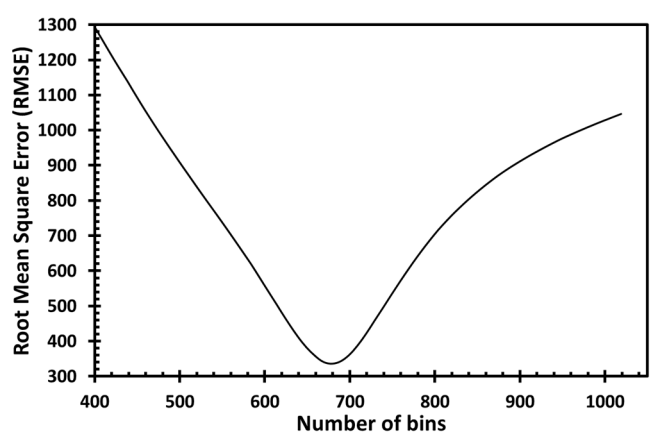

Fig. 4. Root mean square error (RMSE) as a function of number of bins in successive rebinning of simulated spectra. Rebinned spectra were selected at the global minimum of the RMSE.

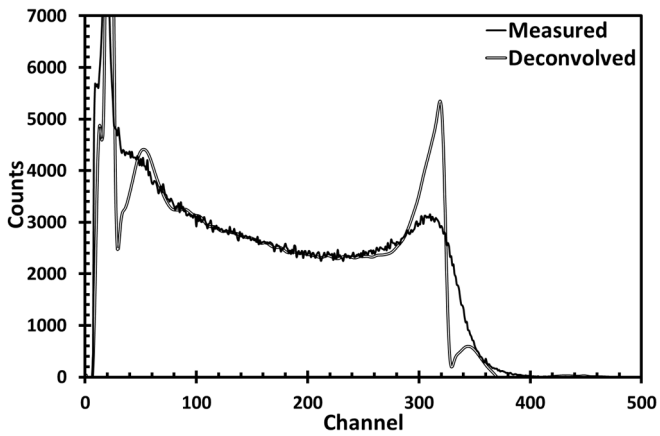

Fig. 5. FFT deconvolved energy spectrum. Few channels difference was observed between the deconvolved spectrum and gain matched simulated energy spectrum that was used used to calculate *R* in (2).

transforms the energy into channel unit. Fig. 5 shows the deconvolved energy with the measured spectrum. The Compton edge is resolved as a sharp edge. It was observed that there is few channels difference between the unfolded Compton edge and the edge in the simulated energy spectrum used for calculating *R* in (2). Some artifacts are evident in the unfolded spectrum that are from the FFT process resulting in low frequency ringing. These artifacts can be suppressed using an iterative SG filter, on the simulated energy spectrum used for *R* calculation. Fig. 6 demonstrates the outcome in the unfolded spectrum as iterative SG is increased. As can be seen, the ringing observed can be suppressed, but the edge sharpness and vertical nature are compromised. However all edges determined with variable number of SG iterations intersect the measured spectrum at the same channel, which is the Compton edge of the measured spectrum.

B. Technique reproducibility

Reproducibility of the implemented technique was tested using variable experimental electronic gain settings. Measurement of a scintillator sample was made at five different amplifier gain settings using Cs-137 source. Gain settings at 3, 4, 5, 6, and 7 were used. Fig. 7 shows the

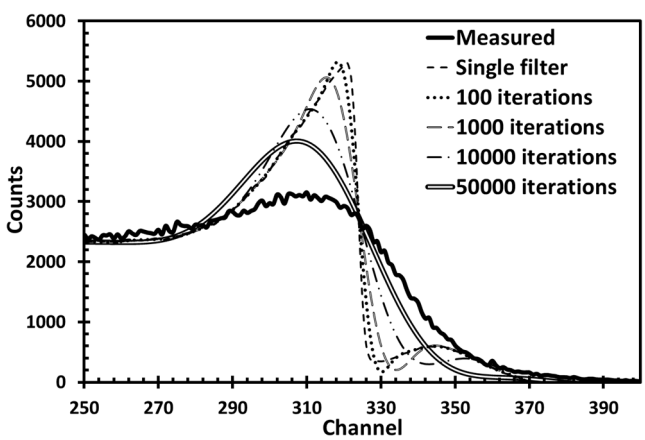

Fig. 6. FFT deconvolved energy spectra. SG filter was used iteratively to suppress ringing in the FFT analysis. The edge sharpness and vertical nature are compromised for increased iterations. The Compton edge is defined at the channel where all edges intersect the measured spectrum.

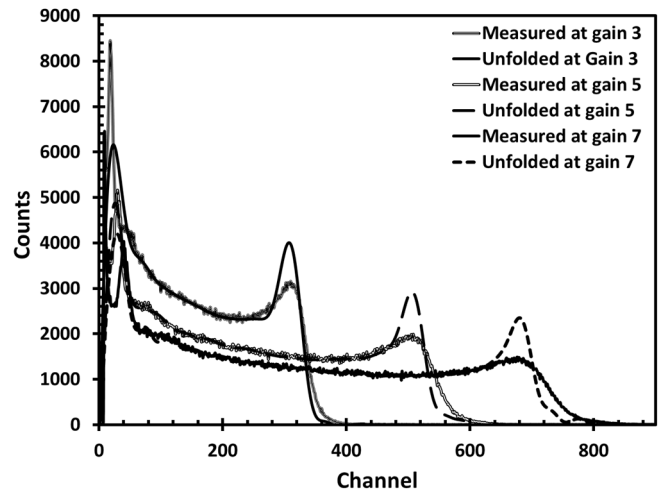

Fig. 7. FFT deconvolved energy spectra at three different experimental amplifier gain settings.

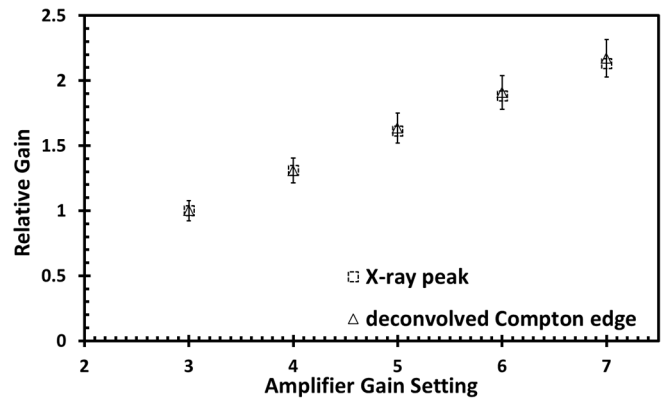

Fig. 7. Relative gains determined using Gaussian fit X-ray peak and deconvolved Compton edge. Amplifier gain settings were incrementally varied from 3 to 7 in the measurement.

measured and unfolded spectra for representative gains at 3, 5, and 7. Before proceeding with the FFT unfolding, the simulated energy spectrum was subjected to the maximum SG iterations used in Fig. 6 to suppress ringing in the unfolded spectra. Ringing observed at the lower and higher channels, shown in Fig. 5, was significantly suppressed. Relative gain in the measured spectrum at each gain setting was determined using the X-ray peak centroids evaluated using Gaussian fitting. Alternatively, the Compton edges determined from the unfolded spectra were also used to determine the relative gain. Fig. 7 shows relative gains as a function of amplifier settings. Good agreement was found between relative gains determined based on the X-ray peaks and deconvolved Compton edges to within 2%.

C. Technique validation

Compton edges determined after implementing the technique outlined in previous section are given in channel unit. It is necessary to confirm that the evaluated edges agree with the expected theoretical values in energy unit thus validate 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

TABLE I

Evaluated Compton edges using NaI(Tl) and BGO detectors

Source (keV)	NaI(Tl)		BGO	
	Evaluated (keV)	%Error	Evaluated (keV)	%Error
Cs137 (478)	473	1.05	480	0.31
Mn54 (638)	632	0.94	641	0.44
Na22 (1061)	1067	0.57	1048	1.23

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.

Calibration of the detectors was 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 followed by FFT unfolding to determine the Compton edges in channel unit. 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 are reasonably proportional in the energy range above 400 keV.

Table 1 tabulates theoretical Compton edges together with those determined using the present technique. Good agreements were found that were within 1% for NaI (Tl) and 1.2% for BGO detector. Assumption of linear response for NaI (Tl) and BGO in the energy range of interest can likely contribute to the slight discrepancy observed. However results are very encouraging and promise the use of the present technique in light yield energy calibration of low resolution detectors such as plastic and liquid scintillators.

IV. Conclusion

In the present work we demonstrated a technique for determination of a Compton edge in a measured plastic scintillator gamma-ray spectrum. MCNP5 simulated pulse height and energy spectra coupled with spectra gain matching and FFT unfolding were instrumental in the Compton edge evaluation. Results obtained were very encouraging and reasonably accurate in localizing the Compton edge. We believe that the present implemented technique can be useful for light yield energy calibration in detectors with low energy resolution such as plastic and liquid scintillators.

Acknowledgments

This work was supported by the Department of Homeland Security-Domestic Nuclear Detection Office (DHS-DNDO) under contract number HSHQDC-13–XB0006-0.

References

[1] G. H. V. Bertrand, F. Sgueera, C. Dehe-Pittance, F. Carrel, R. Coulon, S. Normand, *et al.*, "Influence of bismuth loading in polystyrene-based plastic scintillators for low energy gamma spectroscopy," *J. Mater. Chem. C,* vol. 2, pp. 7304-7312, 2014.

[2] M. Hamel, S. Normand, G. Turk, and S. Darbon, "Highly Lead-loaded red plastic scintillators as an X-ray imaging system for the Laser Mega Joule," *Advancements in Nuclear Instrumentation Measurement Methods and their Applications,* pp. 1-5, 2011.

[3] B. L. Rupert, N. J. Cherepy, B. W. Sturm, R. D. Sanner, and S. A. Payne, "Bismuth-loaded plastic scintillators for gamma-ray spectroscopy," *Europhys. Lett.,* vol. 97, p. 22002, 2012.

[4] M. J. Hwang, Y. J. Kwon, H. J. Kim, J. W. Kwak, S. C. Kim, S. K. Kim, *et al.*, "Development of tin-loaded liquid scintillator for the double beta decay experiment," *Nucl. Instr. Meth. A,* vol. 570, pp. 454-458, 2007.

[5] E.R. Siciliano, J.H. Ely, R.T. Kouzes, J.E. Schweppe, D.M. Strachan, and S.T. Yokuda," Energy calibration of gamma spectra in plastic scintillators using Compton kinematics" *Nucl. Instr. and Meth. Phys. Res. A*, vol. 594, pp. 232-243, 2008.

[6] N. Kudomi, "Energy calibration of plastic scintillators for low energy electrons by using Compton scatterings of γ-rays" *Nucl. Instr. and Meth. Phys. Res. A*, vol. 430, pp. 96-99, 1999

[7] F.B. Brown *et al.*, "MCNP VERSION 5," LA-UR-02-3935 (2002).

[8] R. D. Evans, *The Atomic Nucleus*, McGraw-Hill Inc, New York, 1955, pp. 672–694

[9] J. Allison et al, "Geant4 developments and applications," *IEEE Trans. Nucl. Sci.*, vol. 53, no. 1, pp. 270-278, 2006.

[10] W.H. Press, S.A. Teukolsky, W.T. Vetterling, and B.P. Flannery, "*Numerical Recipes in C*", Cambridge University Press, New York, 1992, pp. 496-558.

[11] Hamamatsu, http://www.hamamatsu.com.

[12] ORTEC, http://www.ortec-online.com/

[13] A. Savitzky and M.J.E. Golay, "Soothing and differentiation of data by simplified least square procedure," *Anal. Chem.,* vol. 36, pp. 1627-1639, 1964.