Benchmarking the Compton Coincidence Technique for Measuring Electron Response Non-Proportionality in Inorganic Scintillators

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Abstract

To study the light yield non-proportionality of inorganic scintillation materials, a Compton coincidence experiment has been designed and implemented. The coincidence technique is used to measure the nearly mono-energetic scintillator electron response by recording events only when energetic electrons are produced by gamma rays that are Compton scattered through a specific angle. This technique provides the ability to accurately determine the light yield non-proportionality of scintillation materials as a function of electron energy while minimizing the potential complicating effects of surface interactions and X-ray escape. To benchmark the Compton Coincidence Technique (CCT), the electron response for NaI(Tl) has been measured for electron energies from 2 keV to 450 keV and compared to previously-published analytical and measured light yield electron responses. These CCT results are believed to be the most accurate NaI(Tl) electron response measurements to date for energies below 20 keV where the light yield non-proportionality is most pronounced.

I. INTRODUCTION

A. Gamma-Ray Response

Scintillation material light yield non-proportionality, sometimes referred to as non-linearity, has typically been characterized by using photopeak centroids from multiple gamma-ray and X-ray sources over the energy range of interest. One disadvantage associated with using gamma rays and X-rays to characterize the non-proportionality of scintillators involves the nature of the interactions within the scintillator. More precisely, the number and energy of electrons produced within a scintillator will vary from one gamma ray to another for equivalent total energy deposition. Consider the total energy absorption of a 662 keV gamma ray in a scintillator. The gamma ray may undergo immediate photoelectric absorption resulting in an electron with a kinetic energy of 662 keV minus the electron binding energy. A gamma ray with the same energy may also Compton scatter one or more times before photoelectric absorption resulting in two or more energetic electrons each with considerably less energy than the first example. As a result, measured gamma-ray and X-ray responses are inherently biased due to light yield non-proportionality with such measurements depending on the size of the scintillator. Thus, gamma-ray and X-ray responses are not ideal for accurately characterizing light yield non-proportionality. Consequently, it has been concluded that measuring the light yield response to electrons will inherently provide a more accurate characterization of light yield non-proportionality and may provide insight into the basic energy transfer mechanisms intrinsic to scintillation materials [2].

B. Electron Response

While the benefit in measuring the electron response has been acknowledged for over three decades, very few studies have reported this scintillator characteristic. Previous methods used to obtain the electron response have relied on two techniques. The first technique involves analytically deconvolving the electron response from measured gamma-ray response data [1,3,4]. This analytical technique requires the accurate determination of electron energy distributions as a function of incident gamma-ray energy. These electron spectra have typically been generated using Monte Carlo simulation. This technique also depends on accurate gamma-ray response data, and thus requires many different gamma-ray and X-ray sources. The second technique is based on direct measurement of electron responses using external electron sources (accelerators or magnetic spectrometers). While there have been attempts to use this second technique to measure electron response non-proportionality [6], these measurements have been particularly subject to complications from surface effects for energies less than 20 keV.

Above 20 keV, both of these techniques seem to be reliable and are in good agreement with each other. However, both techniques rely on data generated using external sources for which there are several factors that effect light yield measurements. Electrons are easily attenuated and have very short path lengths in most inorganic scintillation materials. For example, electrons with energies less than 10 keV have path lengths on the order of a few microns in NaI(Tl). Gamma-ray path lengths in this energy range are not substantially larger. As a result, light yield responses from external sources may easily be influenced by surface effects and X-ray escape. Kaiser, et al. [2] demonstrated that NaI(Tl) light yield is very sensitive to surface treatment below 20 keV. In 1970, Meggit [7] concluded that the effects of surface interaction have impeded the accurate determination of scintillator light yield responses as a function of electron energy below 10 keV and that additional experimental work...
was required. Since that time no progress has been made toward more accurately measuring electron responses at these lower energies.

II. COMPTON COINCIDENCE TECHNIQUE

A. Experimental Technique

A method for measuring inorganic scintillator electron response which addresses the difficulties mentioned above has been proposed [8]. This method is based on a variation of the Compton Spectrometer [9] which utilizes the production of energetic electrons from Compton scattered gamma rays within the scintillator. In this technique, which will be referred to as the Compton Coincidence Technique (CCT), the scintillator to be characterized is exposed to a collimated beam of mono-energetic gamma rays of known energy $h\nu$. A block diagram of the CCT is shown in Fig. 1. Some of the gamma rays will undergo a Compton scatter interaction within the scintillator, transferring energy $E_e$ to an electron. If a Compton scattered gamma ray has no further interaction in the scintillator, it will exit the scintillator at some angle $\theta$ with energy $h\nu'$, given by

$$\frac{h\nu'}{1+(h\nu/m_e c^2)(1-\cos\theta)},$$

where $h\nu$ is initial gamma-ray energy and $m_e c^2$ is the mass equivalent energy of an electron.

Although a continuous spectrum of electron energies is produced due to Compton scattering, a nearly mono-energetic electron response can be resolved by using a second detector in coincidence with the scintillation detector. The second detector, positioned at a specific angle $\theta$ relative to the incident gamma rays, is used to detect the Compton scattered gamma rays. Those events that occur in coincidence in both detectors are stored and analyzed. Using high-purity germanium (HPGe) as the second detector, the Compton electron energy can be accurately calculated by selecting a narrow energy window about $h\nu'$ from the HPGe spectrum and using conservation of energy,

$$E_e = h\nu - h\nu'.$$

This application of the CCT measures the scintillator response to nearly mono-energetic electrons. Since $h\nu'$ and $E_e$ can be varied by changing the angle $\theta$, the measured electron response is not limited to a discrete set of energies but can consist of any number of energies sampled from the continuous spectrum of Compton scattered electrons (Compton continuum).

Complications due to surface interactions experienced by other techniques are avoided when using the CCT because the energetic electrons are produced internally in the scintillator by gamma rays. By using gamma rays with relatively high energy, the gamma-ray interactions (and thus electron production) are spatially distributed throughout the scintillator minimizing interactions near the surface. The effects of X-ray escape are also minimized. Consequently, the CCT provides an assessment of the intrinsic scintillator characteristics that is nearly independent of scintillator geometry and size.

The CCT is also a variation of a procedure that has been used to study liquid scintillator electron responses in the 1-20 keV energy range [10]. However, a significant advantage of the CCT as described in this study is that it minimizes the number of coincidence events that undergo multiple Compton scatter interactions in the scintillator. Using discrete angles and collimation, the ratio of multiple to single Compton interaction events being recorded can be reduced to less than 1 percent [8]. This small fraction of recorded multiple Compton scatters can not be attained with the secondary detector in close proximity to the scintillator.

B. Gamma-ray Source

In this study of the CCT, two monoenergetic gamma-ray sources were used, $^{137}$Cs and $^{99m}$Tc, at a distance of 100 to 150 centimeters away from the scintillator. While the distribution of Compton electron energies theoretically extends down to zero energy, difficulties in resolving low and high angle scattering limited the useful range to angles between approximately 8° and a maximum of 145°. Thus with the 662 keV emission of $^{137}$Cs, the minimum electron energy that could be resolved was approximately 8 keV. The $^{99m}$Tc gamma-ray source, which emits a 140 keV gamma ray, permitted lower energy Compton electrons to be produced at larger angles. The energy range attainable with $^{99m}$Tc also significantly overlapped the $^{137}$Cs energy range, and thus provided a means of comparing results for the two sources. Since $^{99m}$Tc has a half life of approximately 6 hours and acquisition times as long as 24 hours were required before replacing the $^{99m}$Tc, high initial activities (150 millicuries) were used with attenuators placed next to the source (see Fig. 1). Every
whether a valid signal had occurred in the gated scintillator ADC). This method requires the HPGe detector count rate at the energy of interest to be less than 100 counts per second which is easily achieved in the CCT.

Since data collection typically consisted of several hours or more, gain drifts in both detectors were monitored closely and quantified by periodically recording both the HPGe anti-coincidence and total scintillator pulse height spectra which included photopeaks from either the mono-energetic gamma-ray source or an additional external source placed in close proximity to each detector. Measuring the relative positions of specific photopeaks as a function of time served as a method of quantifying system gain drift. All coincidence data were recorded in a sequential list mode with a time stamp, thus permitting post-acquisition analysis to correct for any gain drift.

Accurate electron response measurements also require a PMT and other electronics with as near a linear response as possible or with known non-linearity characteristics. The linearity of the PMT and electronics used in this study were characterized using a variation of the dual source technique. The results were not used to correct data but were propagated into reported uncertainties contributing to less than 0.1 percent.

The NaI(Tl) scintillator used in this study was a 2.54 by 2.54 cm cylindrical crystal. The scintillator was mounted on a Hamamatsu R329-02 photomultiplier tube with a tapered voltage divider to maximize the linear range of the PMT. The secondary detector consisted of a HPGe detector with a beryllium end-window.

### C. Instrumentation

A schematic of the nuclear instrumentation used in the CCT is shown in Fig. 2. The coincidence electronics included a time-to-amplitude convertor/single channel analyzer (TAC/SCA) with a coincidence resolving time of approximately 150 nanoseconds. The TAC/SCA was used to gate the scintillator analog-to-digital convertor (ADC). A multichannel analyzer card in the computer was used to record the entire pulse height spectrum from the scintillator. It should be noted that the HPGe ADC was not gated with the TAC/SCA. Instead, both ADCs were used in a synchronous mode that permitted each ADC to inhibit the other ADC after peak detect had occurred. This operating mode permitted the computer to identify and store pulse height data for the HPGe detector as either coincidence or anti-coincidence (based on whether a valid signal had occurred in the gated scintillator ADC). This method requires the HPGe detector count rate at the energy of interest to be less than 100 counts per second which is easily achieved in the CCT.

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### III. EXPERIMENTAL RESULTS

#### A. NaI(Tl)

Since NaI(Tl) is the most extensively studied and best characterized inorganic scintillator to date, it was chosen for benchmarking the CCT. NaI(Tl) is generally regarded as having good light yield proportionality, however it suffers from non-proportionality for energy depositions below 200 keV. During the 1950s and 1960s, significant efforts were made to quantify NaI(Tl) light yield non-proportionality as a function of electron energy with good agreement for electron energies above 20 keV. Prescott and Narayan [5] present a summary of these efforts.

#### B. Analysis

NaI(Tl) pulses were included in the CCT analysis only if the coincidence energy pulse from the HPGe detector fell within a specified energy window. The width of this energy window depended on the scattered gamma-ray energy and the coincidence efficiency, varying between 500 eV (for low angle scatters) to 5 keV (for high angle scatters using $^{137}$Cs). This energy discrimination typically reduced chance coincidences to less than 1 in 1000.
The resulting coincidence pulse height spectrum for the NaI(Tl) scintillator was a Gaussian shaped peak. To determine the centroid, the top half of the peak was fit to a Gaussian curve using Chi-squared minimization. The electron response was determined by comparing the average light yield for the electron energy in the NaI(Tl), to a photoelectric peak in the NaI(Tl). The CCT results are reported as relative light yield per electron energy and are tabulated in Tables 1 and 2 for $^{137}$Cs and $^{99m}$Tc, respectively. The ideal relative scintillator light yield response ($LE_\varepsilon$) is a constant for all energy depositions.

C. Comparison to other Electron Responses

The relative NaI(Tl) light yield response to electrons measured with the CCT is compared to the results of three previous studies [1,4,6] as shown in Fig. 3. The CCT results and other curves are arbitrarily normalized at 45 keV. Uncertainties typically increase with decreasing electron energy and are shown for those measurements where error bars are larger than the symbol's size. Error bars for results above 10 keV are less than 1 percent and include the propagation of uncertainties due to counting statistics, centroid calculations, system linearity, and HPGe calibration.

The work by Porter, et al. [6] illustrated in Fig. 3 is based on direct measurements using external electrons and is in good agreement with the results by Zerby, et al. [1] above 20 keV, extending to 1000 keV. While Porter's measurements above 20 keV are assumed to be accurate, below this energy the results are subject to surface effects resulting in measured relative light yields much smaller than are intrinsic to NaI(Tl).

Both the work of Zerby, et al. and Hill and Collinson [4] are based on analytical calculations as briefly described above. It has been reported that the derived electron responses of Hill and Collinson may represent the most accurate responses available for electron energies below 10 keV due to their accounting for L-shell excitation [5]. However, the subsequent detection of 0.87 keV events in NaI(Tl) by McCain and Smith [11] indicates that these calculated light yields for energies less than 10 keV are likely too low. The results obtained with the CCT in this study seem to support this. Miggitt [7] has attempted to correct Collinson and Hill's responses to account for surface effects and X-ray escape. These corrected results have a larger light yield response for lower energy electrons and more closely resemble the CCT results.

IV. DISCUSSION OF RESULTS

A. Light Yield Response above 20 keV

Above 20 keV, there is excellent agreement between the CCT data and previously measured light yield responses for NaI(Tl). The calculations of Zerby, et al. and the direct measurements of Porter, et al. both verify the results obtained with the CCT and agree to within 1 percent for energies above 20 keV when normalized to each other at 45 keV. These results indicate that the CCT is accurately characterizing the electron response non-proportionality of NaI(Tl).

The 450 keV limit of the CCT in this study may be further extended by using a higher energy gamma-ray source with energy higher than that of $^{137}$Cs. The maximum electron energy that may be measured is only limited by the Compton continuum produced from the gamma-ray source along with any physical constraints in the scattering angle.

B. Light Yield Response below 20 keV

The CCT measurements result in a maximum light yield response around 10 keV, similar to the results of the other work shown in Fig. 3. However, the CCT indicates a slightly larger light yield response near the maximum, then follows the trend calculated by Zerby, et al. [11] at lower electron energies (less than 10 keV).

| Table 1 | Electron Response for NaI(Tl) using the Compton Coincidence Technique and $^{137}$Cs (661.66 keV). |
| Electron Energy $E_\varepsilon$ (keV) | Relative Light Yield per Energy of $LE_\varepsilon$ | Uncertainty of $LE_\varepsilon$ (\sigma) |
| 444.0 | 1.036 | (0.001) |
| 311.6 | 1.049 | (0.002) |
| 218.1 | 1.061 | (0.002) |
| 151.4 | 1.084 | (0.003) |
| 107.0 | 1.106 | (0.002) |
| 74.4 | 1.137 | (0.002) |
| 49.7 | 1.170 | (0.002) |
| 35.0 | 1.199 | (0.005) |
| 23.6 | 1.232 | (0.003) |
| 16.1 | 1.260 | (0.005) |
| 10.8 | 1.262 | (0.004) |
| 7.93 | 1.250 | (0.008) |

| Table 2 | Electron Response for NaI(Tl) using the Compton Coincidence Technique and $^{99m}$Tc (140.51 keV). |
| Electron Energy $E_\varepsilon$ (keV) | Relative Light Yield per Energy of $LE_\varepsilon$ | Uncertainty of $LE_\varepsilon$ (\sigma) |
| 25.4 | 1.218 | (0.006) |
| 17.2 | 1.246 | (0.004) |
| 11.8 | 1.270 | (0.004) |
| 8.24 | 1.265 | (0.005) |
| 5.83 | 1.242 | (0.008) |
| 4.07 | 1.212 | (0.010) |
| 3.05 | 1.181 | (0.011) |
| 2.03 | 1.138 | (0.021) |
The lower limit of electron energy for the CCT, as configured in this study, was found to be approximately 2 keV, primarily due to the coincidence electronics for the NaI(Tl) detector. Difficulties were experienced in discerning coincidence events in the NaI(Tl) detector at 1 keV. A loss of timing signals in this energy range resulted in biasing data, presenting error in the analysis for energies below 2 keV. Light yield responses in this energy region may be achievable with different coincidence electronics and a PMT designed for single photoelectron counting.

C. Special Considerations

The accuracy of the CCT results was found to be sensitive at electron energies below 10 keV. Most significantly, an accurate calibration of the HPGe detector was required to minimize errors in the light yield response. The CCT analysis was very sensitive to small differences in HPGe energy calculations with the effects of calibration source location often being more significant than statistical uncertainties. It is believed that such issues have been sufficiently addressed and that the CCT data presented in Fig. 3 represents the most accurate NaI(Tl) electron response available.

Low coincidence rates required several hours of data collection for each angle when using the $^{137}$Cs source. Even lower coincidence rates were experienced with the $^{99m}$Tc source which emits a 140 keV gamma ray. Due to the high probability of photoelectric absorption in NaI(Tl) for 140 keV gamma rays, data collection required as much as 24 to 48 hours to obtain a statistically significant number of events for one angle. The low coincidence rates are also partly due to the relatively large NaI(Tl) crystal being used. Coincidence rates would likely be improved by using a smaller scintillation crystal and a more efficient (larger) HPGe detector.

Another consideration in the CCT measurements is the effect of electron binding energy. Compton scattering with a bound electron will result in a Compton electron with slightly less energy than is calculated by Eqn. (2). However, unlike photoelectric absorption, Compton scattering involves all electrons in the scintillator medium, thus resulting in a small average binding energy. At this time, it is believed that binding energy has a negligible effect on CCT results.

V. SUMMARY AND CONCLUSIONS

The Compton Coincidence Technique (CCT) has been demonstrated to accurately characterize the intrinsic light yield non-proportionality of NaI(Tl) as a function of electron energy. Consequently, the light yield response to electrons of other inorganic scintillators may now be confidently measured using the CCT.

Several distinct advantages are evident when using the CCT. A continuous distribution of Compton electron energies are available and only a few gamma-ray sources are needed to cover a broad range of electron energies. Interactions at the scintillator surface are minimized since the energetic electrons are produced internally. As a result, electron responses can be measured at lower energies with the CCT than has been possible with other techniques that have relied on external
sources. The successful application of the CCT could significantly impact the assessment of new scintillators and further the study of the scintillation process in both well-characterized and new scintillators. Furthermore, the CCT can be used for the assessment of scintillator intrinsic energy resolution along with the development of more accurate scintillation detector response functions.

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VII. REFERENCES


