Results from an investigation into the physical origins of nonproportionality in CsI(Tl)

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Abstract

The relative scintillation response per energy deposited by Compton electrons or, nonproportionality, has traditionally been considered an intrinsic scintillator property. However, such an interpretation is inconsistent with recent results that show nonproportionality to depend on external factors such as shaping time, temperature and supplier. Apparently, at least some of the overall nonproportionality has an extrinsic origin. In this work we describe the results from a suite of measurements designed to test the hypothesis that nonproportionality in CsI(Tl) material has an extrinsic component that correlates with impurity levels. Our choice of material was motivated by the excellent energy resolution observed in one bulk crystal (6.4 %) - a marked departure from that measured with conventional CsI(Tl) stock (8 to 8.5 %). Six bulk CsI(Tl) crystals were procured and diced into 44 wafers. Using x-ray fluorescence techniques no conclusive evidence for impurities were found in any of the wafers at the 1 to 50 ppm level. One crystal exhibited a distinct correlation between energy resolution, decay lifetimes, non-proportionality and a very low level of Tl doping.

nonproportionality; CsI(Tl); impurities; x-ray fluorescence

1.0 Introduction

The relative scintillation response per energy deposited by Compton electrons or, nonproportionality, has traditionally been considered an intrinsic scintillator property. However, support for this interpretation is increasingly untenable in light of measurements that have shown nonproportionality to depend on external factors, such as shaping time and temperature[1]. Consequently, notions on the origin of nonproportionality have evolved to the present moment, where it is gradually acknowledged that at least some of the overall nonproportionality is not an intrinsic scintillator property. The primary goal of this work was to observe correlations between nonproportionality and impurities (an extrinsic property) at the bulk crystal level. A secondary (and more subtle) goal was simultaneously pursued to

explore whether sub-crystal variation in impurity levels contributed to bulk crystal nonproportionality. Our choice of CsI(Tl) material was driven by the excellent resolution seen with one crystal (~ 6.4%) - a marked departure from that documented for conventional CsI(Tl) stock (8 to 8.5%). This disparity suggests that should nonproportionality originate from impurities and should these impurities be controllable, then there exists the exciting possibility that the resolution of CsI(Tl) could be engineered to achieve 2% resolution, i.e., comparable to $LaBr_3$. Since CsI(Tl) is very widely disseminated, such an improvement could potentially revolutionize nonproliferation efforts as well as nuclear, particle and medical physics.

2.0 Measurements

To test the hypothesis that nonproportionality is due, in part, to impurities we obtained six bulk crystals from five growers worldwide. (To preserve anonymity concerning growth methods these crystals are subsequently referred to by suppliers designations A, B, C, D, D' and E, with the redundant use of D and D' denoting two crystals provided by the same supplier). Guided by ray trace simulations (Figure 1) we converged on a wafer geometry that would simultaneously provide good light collection, minimize a geometric contribution to energy resolution and be readily wrapped and positionable. The six bulk crystals were then each diced into 4 to 9 4.5 mm x 1 in wafers to explore whether wafer variability contributes significantly to overall crystal performance. A dedicated mounting rig (Figure 2) was designed and built to ensure highly reproducible coupling between the wafers and PMT surface. Nonproportionality, energy resolution, decay lifetimes measurements and x-ray fluorescence (XRF) measurements were performed on each of the 44 samples using one mounting rig.

2.1 Nonproportionality

A pair of 5 μCi ^{137}Cs sources was used to excite the CsI(Tl) wafers optically coupled to the horizontal PMT (Figure 2). An XIA Pixie-4 spectrometer recorded coincidence events comprised of the energy of a Compton-scattered photon measured in the germanium detector and that of the energy from light generated by the recoiling electron measured by the PMT. (The sum of these energies of these coincident events is equal to that of the source energy - 662 keV. The energy of the recoiling electron is derived by subtracting the energy deposited in the germanium detector from the source energy.) A 1000 ns coincidence window allowed sufficient random coincidences to pass such that energy resolution measurements could be extracted directly from the data sets without need for a separate calibration run. Data were written to disk in 10 minute intervals and corrected offline for PMT and

germanium gain drift. Pixie-4 software designed for phoswich detector applications [2] was used to construct calibrated 2-D coincidence matrices formed by co-adding ~25-30 data files collectively comprising $2x10^6$ events. To keep bin-to-bin fluctuation small these sparse matrices were compressed by a factor of ~ 50, culminating finally in a 600 x 600 bin 2-D matrix with a resolution of 1.6 keV/bin. 1-D matrices were constructed by projecting along the axis representing the recoiling electron energy. Local maxima in these projections were fit with a Gaussian and the centroid of the Gaussian fit recorded.

2.2 Decay Lifetimes

A Golden Engineering XR-200 x-ray generator with a peak output around 40 keV and 55 ns pulse width was used to excite the wafers using the same mechanical mounting rig shown in Figure 2. Pixie-4 DSP code was modified to allow for high frequency sampling of short decay components and lower frequency sampling of longer decay components to accommodate (unknown) decay components waveforms as long in duration as ~ 100 us. The waveforms were fit with a triple exponential function to recover the fast, intermediate and slow decay components (0.6 μs, 3.3 μs and 10 μs, respectively).

2.3 XRF

All 44 wafers were sent to Matrix Metrologies (El Granada, Ca) for energy dispersive XRF measurements. Six of these wafers (one from each bulk crystal) were sent to Material Characterization Services (Austin, Tx) for high precision wavelength dispersive XRF measurements.

3.0 Results

Results from the nonproportionality, energy resolution and decay lifetime measurements performed on the individual wafers are grouped by supplier in Figures 3 and 4. Results from the XRF measurements are contained in Table 1. No statistically significant variation in nonproportionality, energy resolution or decay lifetimes is observed among wafers cut from the same bulk crystal. This uniformity prompted an alternative analysis wherein each wafer from the same bulk crystal is treated as an independent sample of bulk crystal properties. With this approach, individual nonproportionality, energy resolution and decay lifetime measurements are statistically combined to extract average properties for each bulk crystal. Figure 4 also contains the results of this averaging procedure for the decay lifetimes, while Figure 5 separately contains the combined non-proportionality and energy resolution results. When examined at the bulk crystal level it is manifestly evident that five of the six crystals are essentially indistinguishable from one another (within measurement error). In a similar vein, the crystal from

supplier D is clearly an anomaly with the poorest energy resolution (~ 9.0%) and shortest decay lifetimes of any of the other five crystals. Presumably, these differences are explained by the results of the EDXRF measurements (Table 1), which reveal no evidence of Tl (or Na) in this same crystal. (The non-detection of Tl (or Na) is somewhat surprising since the general shape of its nonproportionality curve is still reminiscent of CsI(Tl).) Presumably, the doping level in the crystal from supplier D is far from optimal for the wafer size (before cutting this crystal showed the best energy resolution ~6.4%). The Tl doping level variations from ~150 ppm to ~500 ppm (WDXRF results) exhibited by the other crystals do not seem to have an effect on non-proportionality or energy resolution.

4.0 Conclusions

Modern CsI(Tl) growth techniques methods are apparently capable of consistently producing very high purity crystal stock, with even the small (~ 10%) variation measured among NaI(Tl) crystals from different suppliers[3] apparently suppressed in CsI(Tl). There is little evidence in the work described herein to support the proposal that CsI(Tl) nonproportionality can be engineered via judicious control of impurities above the 50 ppm level (50 ppm for Na down to 1 ppm or less for Tl and heavy impurities) to improve its energy resolution. However, it is possible that impurities and/or the Tl dopant are being controlled by supplier D at a very low level to achieve the excellent resolution measured in the bulk crystal. Resolution of this conundrum must await analysis by more precise methods, including particle induced x-ray emission, neutron activation or mass spectrometry.

Acknowledgments

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References

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