Growth of Detector-grade CsI:Tl Scintillator Crystals by Gradient Freeze Technique

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Introduction

Tl doped CsI single crystal is old and well known scintillator material [1]. It has good scintillation efficiency and is less hygroscopic and less brittle than NaI. This material could not achieve its full potential in the past due to the unavailability of a matching PMT, however, use of PIN photodiodes, which show good efficiency at longer wavelengths and compact in size, has renewed the interest in the CsI(Tl) crystals. Also, since CsI:Tl shows different decay times for different charge particles, pulse-shape discrimination techniques can be effectively used for the particle identification [2].

Single crystals of CsI can be grown by Bridgeman as well as Czochralski methods. Though, the crystal growth using the Bridgman method is not a problem, sticking of the crystal with crucible wall and hence recovery of the crystal after the growth and thermal and mechanical stresses generated therein are important issues of considerable interest [3]. In this paper we are reporting the growth of CsI single crystals in carbon coated silica crucibles by a gradient freeze technique. Gamma-ray detectors were fabricated and characterized to study the effect of growth conditions on the quality of crystals.

Experimental

Crystal growth furnace was designed to have a positive temperature gradient from top to bottom. Fig. 1 shows the gradient of the furnace with a blank crucible. To see the actual temperature profile in the melt, KCl that can be heated in air and has a heat capacity approximately equal to CsI and melts at 780°C, was taken in the silica crucible and a K-type thermocouple enclosed in a thin silica tube was inserted in the melt. The temperature profile in the melt is also shown in Fig.1.

Silica crucible of one inch diameter and having conical bottom were taken for growth. For carbon coating crucible was first cleaned by a 10% HF solution for few minutes. Cleaned crucible was transferred to a horizontal tubular furnace and heated upto 900 C, then hexane was flown in the furnace which on cracking deposited carbon on the crucible wall. As coated crucible was then transferred in a vacuum furnace and annealed at 1100 C.

High pure (99.995%) and dry CsI and TlI (0.2 mole%) were taken in the carbon coated crucible and sealed under 10^{-3} mbar running vacuum at 200°C. Crucible was then put inside the furnace on a stationary growth station and temperature of the furnace was raised so that the the bottom of the crucible was at 640°C temperature (20°C higher than the melting temperature of the CsI). Melt was kept at that temperature for 4 h and then cooling was started at a rate of 4°C/h.

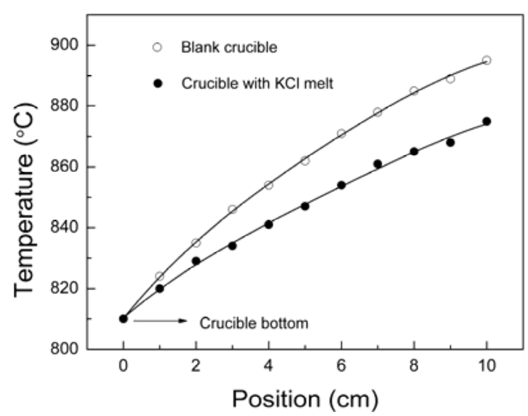

Fig.1: Temperature profile inside the crucible (○) without melt and (●) with melt.

25 mm Φ x 20 mm L cylinder was cut from crystal ingot and polished as shown in figure 2. As processed crystal was then wrapped with 10 layers of Teflon tape leaving one face open to connect with PMT. Gamma spectra of Cs^{137}, Cs^{134}, Co^{60}, Na^{22} and Co^{57} were

recorded using detector (CsI:Tl+PMT+preamplifier), spectroscopic amplifier and multi-channel analyzer (8k MCA).

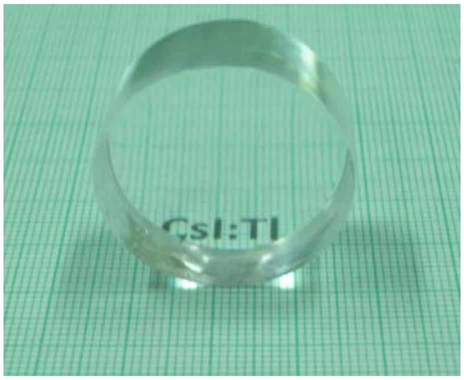

Fig.2: Photograph of polished crystal cylinder.

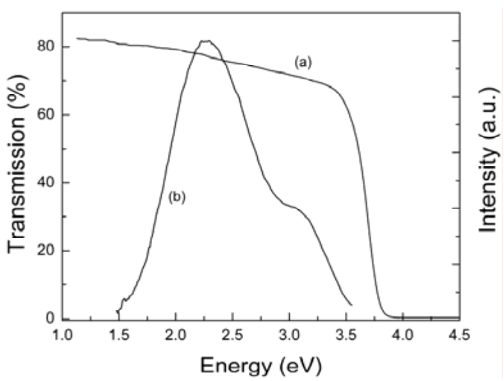

Fig.3: (a) Transmission and (b) luminescence spectra of 0.2% Tl doped CsI crystal. (Sample thickness for transmission experiment was 20 mm).

Results and discussion

Crystal ingots were visibly clear of any carbon particles and were highly transparent. As no part of the furnace or crucible was moving or vibrating the probability of peeling off of the carbon film from crucible walls was the minimum. Transmission and luminescence spectra of the crystal are shown in Fig.3. The spectra do not show extra peaks related to carbonate or any other impurities. This observation supports that the carbon coating was of good quality without any oxygen contamination or impurities.

Linearity of the pulse height response of the detector was checked up to 1332 keV gamma rays. Calibration curve for the detector is shown in Fig.4 and a typical Cs^{137} gamma spectra is also depicted in the same figure showing an energy resolution of ~7.8% at 662 keV which is comparable to or better than reported values [4]. Optimization of light collection at the PMT photocathode was necessary to get the best results. To achieve this nearly 10 layers of Teflon, which has reflectivity better than 98%, was wrapped around the crystal and optical grease was used to couple the crystal with the PMT avoiding air bubbles trapped in between.

Thus it is clear that high quality single crystal of Tl doped CsI can be grown using the gradient freeze technique as described above.

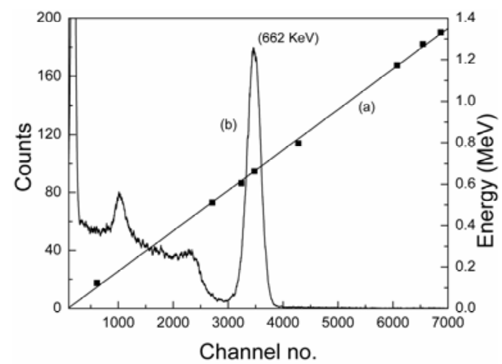

Fig.4: (a) Callibration curve (■) (Energy Vs channel no.) and (b) a typical Cs^{137} gamma spectra.

Conclusions

High quality single crystals of 0.2 mole % Tl doped CsI were grown by a gradient freeze technique. Carbon coated crucibles of fused silica were used to avoid sticking of crystal with the crucible wall. The grown crystals were extracted from the crucible without involving an inversion process thereby avoiding thermal and mechanical stresses. The transmission and luminescence measurements do not have adverse effects of carbon coating on the crystal quality. Using these crystals γ-ray detectors were fabricated which show good linearity and 7.8% resolution at 662 keV.

References

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