Growth of Tl doped CsI and NaI single crystals in a modified furnace based on Bridgman technique

S.G. Singh, D.G. Desai, A.K. Singh, M. Tyagi, S. Sen and S.C. Gadkari*
Technical Physics Division, Bhabha Atomic Research Centre, Mumbai - 400085, INDIA
** email: gadkari@barc.gov.in*

Introduction

Doped halides are the oldest and most widely used scintillator materials to date. Some of these (NaI:Tl, CsI:Tl, CsI:Na etc) are nearly 60 years old but active research on these materials to improve their scintillation properties is still continued. Single crystals of halides are generally grown by Kyropoulos, Bridgman and Czochralski techniques. Among these techniques the Bridgman technique is useful to grow small to medium size crystals (10 mm – 80 mm diameter).

The growth of crystals using the Bridgman method is relatively simple, however, sticking of the grown crystal to the crucible wall and hence its recovery after the growth and thermal/ mechanical stresses generated are important issues of considerable interest [1]. Luminescence and scintillation properties of grown crystals depend on various crystal growth parameters viz starting material preparation, growth process and post growth treatments [2]. In the present paper we report the growth of good quality single crystals of Tl doped CsI and NaI using a modified vertical Bridgman crystal growth technique. Gamma-ray detectors were fabricated and characterized to check the quality of the grown crystals.

Experimental

The furnace employed for the crystal growth consists of four independently controlled resistive heating elements. The top isothermal zone consisting of two heating elements is separated from the bottom isothermal zone by a 50 mm thick baffle (adiabatic zone). A maximum 20^{o}C/cm temperature gradient was achieved in the adiabatic zone. The axial and radial temperature profiles of the furnace are shown in Fig.1. An especially designed silica glass crucible containing the material is loaded in the upper zone. The temperatures of all the zones are raised in such a manner such that the temperature of the upper zones is 50°C above the melting temperature (MP) of the material while the lower zone temperature is 50°C lower than the MP. Once the material inside the crucible melts completely it is allowed to thermalize for 4 h. The crucible is then lowered at a typical rate of 0.5-2 mm/h to the bottom zone of the furnace through the temperature gradient (adiabatic zone) for a complete solidification of the melt into single crystal. After completion of single crystal growth the crucible is locked using a locking mechanism inside the furnace and the furnace together with the crucible is inverted (rotated through 180 degree). Now the temperature of the zone 3 containing the grown crystal is raised so that it becomes slightly higher than the MP of the material and the grown crystal slides down in the lower zone of the crucible that has a slightly larger diameter. Temperatures of the all the zones are then lowered quickly the same temperature (MP-50°C) to achieve a uniform temperature in the furnace. The crystal is annealed at this temperature for 4 h. Finally the furnace is cooled down to room temperature at a uniform rate of 10-30ºC/h. Afterwards the silica glass crucible is cut open to retrieve the grown crystal.

Gamma detectors of NaI:Tl and CsI:Tl (typical size 45 mm diameter and 45 mm length) were fabricated and tested for the scintillation performance of the grown crystals using a PMT (ADIT: B51D01S) based readout. A scintillator cube (18x18x18 mm^3) cut from a CsI:Tl crystal was further tested using a photodiode (PD: Hamamatsu S3204-08) read out. About 5 to 7 layers of Teflon tape (80 micron thick) were used as reflector in all the experiments.

Results and Discussion

The benefit of the present method is that during the cooling, the grown crystal is not in contact with the crucible wall and therefore it is not subject to any mechanical stress. Over 60 single crystals of CsI:Tl (0.2mol%) and NaI:Tl (0.1mol%), 50 mm diameter and 50 mm length have been successfully grown by this technique with almost 100% yield. All the grown crystals were transparent and crack-free (Fig.2). Though some NaI:Tl crystals were cracked slightly at the bottom, the crack did not propagate in the bulk and could be removed during the processing of the crystal into a detector.

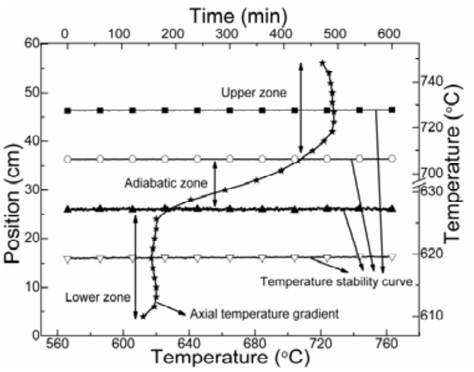

Fig. 1 Axial temperature gradient and temperature stability plot of the furnace.

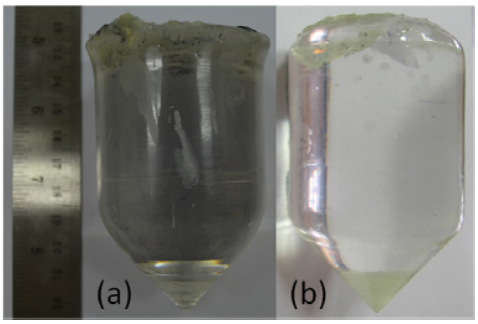

Fig. 2 Photographs of (a) CsI:Tl (0.2 mol%) (b) NaI:Tl (0.1 mol%) single crystals.

The NaI:Tl crystal was tested employing a 1 μs shaping time. The ^{137}Cs and ^{60}Co were used as gamma sources. A typical gamma spectrum of ^{60}Co is shown in Fig.3.The typical resolution at 662 kev was found to be 6.8 ±0.2% while at 1332 keV the resolution was 4.8 ±0.2%. These values are similar to the best commercially available NaI:Tl detectors. The resolution of the detector can be further improved using a proper reflector other than Teflon. The CsI:Tl was tested using a photodiode as well as PMT read outs. A typical gamma spectrum of ^{60}Co measured using CsI:Tl-photo-diode combination is shown in Fig.4.

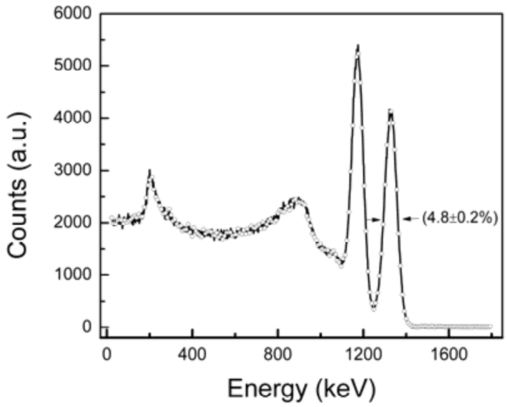

Fig. 3 Typical gamma spectra of ^{60}Co recorded using a 45 mm dia x 45 mm length NaI:Tl.

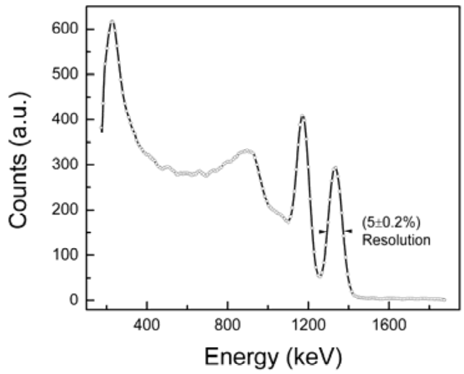

Fig. 4 Typical gamma spectra of ^{60}Co recorded using a 18x18x18mm^3 length CsI:Tl and a PIN diode.

References

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