Growth of SrI_2:Eu single crystals using in-situ filtration of melts

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Introduction

SrI_2:Eu, an old scintillator [1], was recently re-discovered as one of the brilliant scintillator because of advancement in crystal growth, material dehydration and material handling. Photo-yield of SrI_2:Eu is estimated by various groups to be between 80000 to 120000 photon/MeV with a peak wavelength at around 435 nm. It also shows excellent non-proportionality in photo-yield particularly at lower energies due to extremely efficient capture of excitation on Eu^{2+} sites, making it less dependent on excitation density [2]. These desirable properties lead to a resolution better than 3% at 662 keV [3]. However the scintillation properties SrI_2:Eu highly depend on material purity and oxygen/moisture content in the material.

In this paper we report the growth of SrI_2:Eu single crystal using the Bridgman crystal growth technique. A specialy designed silica crucible was used for the growth and melt filtering simultaneously. The grown crystals were characterized for their scintillation performance.

Experimental

For the single crystal growth, 99.99% pure SrI_2 and EuI_2 (Lanhit, Russia) were taken in a specially designed silica crucible (Fig. 1a). The crucible had two parts; the lower part of the crucible had a grain selector and was separated from the upper part by a quartz filter (20-40 micron pour size). Initially the charge is loaded in the upper part of the crucible inside a glove box (MBRAUN make MB-200B) having moisture and oxygen levels below 0.1 ppm. The crucible was then transferred to a dehydration unit and the material was dehydrated at 200°C for 5 h under $5x10^{-4}$ mbar pressure. Thereafter the crucible was sealed under running vacuum by an LPG torch.

Crucible was then transferred to an indigenously designed Bridgeman crystal growth system consisting of three zones. The crucible was first heated to 550°C in the upper zone and was kept there for 24 h. During this time the melt got filtered through the quartz filter and collected in the lower part of the crucible. Afterwards the crucible was lowered to the third zone of the furnace (kept at 400°C) through a gradient of about 20°C/cm (second zone) at the rate of 0.5 mm/h.

For the scintillation characterization a sample of 5x5x5 mm^3 was cut from the ingots and was processed and coupled to a PMT (R6094). For recording gamma spectra the PMT output was processed by a preamplifier (CAEN A1424), shaping amplifier (ECIL PA4903N) and Tukan-8k-USB MCA. A 10 micro-second Gaussian pulse shaping was used in all experiments.

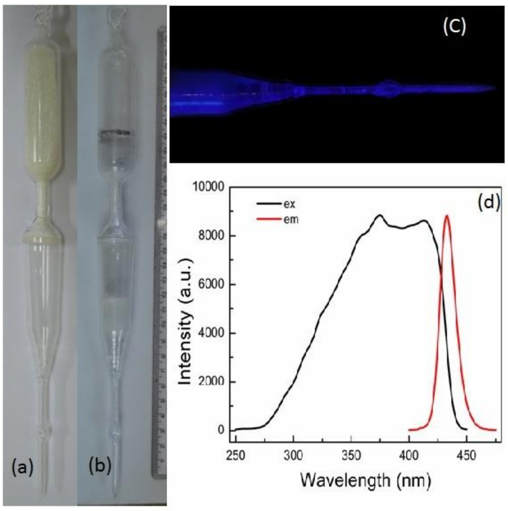

Fig. 1 Crucible photograph (a) with material before growth, (b) after growth (c) under UV and (d) Photoluminescence spectra of crystal.

Results and discussion

Crack-free transparent crystals of 10 mm diameter and 15 mm length were grown successfully employing a melt filtration technique. There was no visible inclusion in the bulk of the grown crystal. All the insoluble material/impurities (10% of initial material consisting large amount of black and brownish particle) got filtered and remained in the upper part of the crucible. The visual inspection shows that the grain selector played an important role. While initially at the bottom many small grains grew only one grain was allowed to grow further. The photoluminescence (PL) spectra of the crystal show a relatively narrow emission peak around 432 nm associated with a broad and overlapping excitation spectrum (Fig. 1 d).

The decay profile of SrI_2:Eu in response of ^{137}Cs excitation is shown in Fig. 2. The average decay time was 900 ns. Considering the slow scintillation response, a 10 micro-second shaping time was used to ensure full collection of the scintillation during gamma spectrometery.

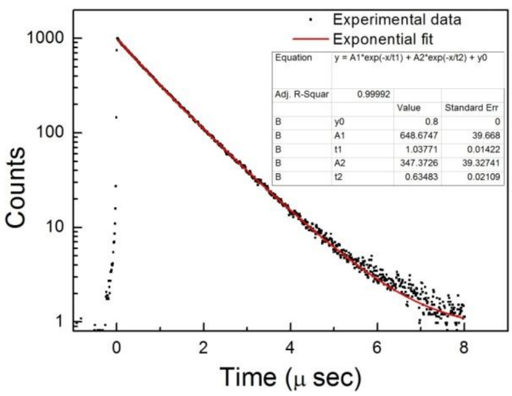

Fig. 2 Decay profile of SrI_2:Eu (5 mol%).

Fig. 3 shows a typical gamma spectrum of ^{137}Cs recorded using 5x5x5 mm^3 sample. The calculated resolution at 662 keV is 3.8%. To assess the non-proportional response of the grown crystal various radio nuclides were used to cover the range from 14 keV to 1332 keV. The normalized response of the scintillator is shown in Fig.4. The recorded response is in agreement with the reported values in literature [2].

In summary, better optical quality of grown crystal, good non-proportional response and consequent better resolution indicate that the melt filtration was effective in removing various impurities form the material during the growth.

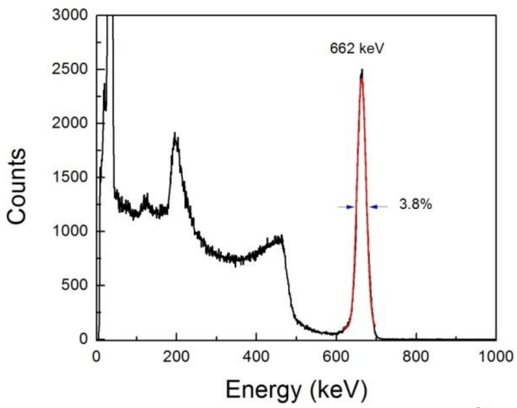

Fig. 3 A typical gamma spectrum of ^{137}Cs recorded by SrI_2:Eu crystal.

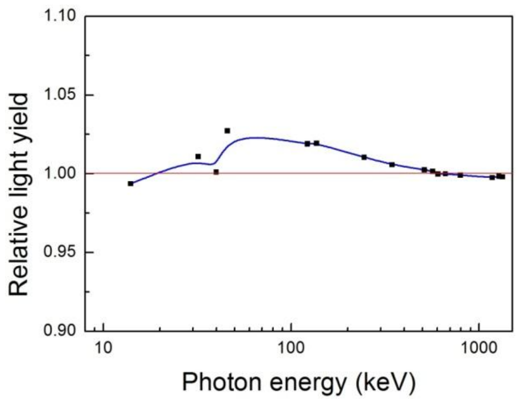

Fig. 4 Non-proportionality of scintillation response of SrI_2:Eu.

References

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[2] Cherepy *et al*, Appl. Phys. Lett. **92**, 083508 (2008).

[3] Van Loef *et al*, *IEEE Trans. Nucl. Sci.*, **56**, 869 (2009).