



Performance characteristics of thermal neutron detectors based on $\text{Li}_6\text{Y}(\text{BO}_3)_3\text{:Ce}$ single crystals



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ABSTRACT

Crack-free single crystals of Ce doped $\text{Li}_6\text{Y}(\text{BO}_3)_3$ (LYBO:Ce) have been grown using the Czochralski technique. Grown crystals were characterized for their optical and scintillation characteristics to explore their potential as neutron detectors. Scintillator detectors based on LYBO:Ce crystal were used successfully to record the pulse height spectra from various neutron sources in the flux range from $10 \text{ n/cm}^2/\text{s}$ to $10^7 \text{ n/cm}^2/\text{s}$. The detection efficiency for thermal neutrons was found to be over 80% for a 2 mm thick LYBO:Ce crystal. The scintillation decay times measured for neutron and gamma radiations were about 27 ns and 49 ns, respectively.

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1. Introduction

The need for neutron detectors has greatly increased with the increase in the number of high flux neutron sources, threat from nuclear materials, neutron imaging and many other applications [1]. The neutron imaging has also emerged as an important tool for homeland security. Currently, ^3He and BF_3 gas filled proportional counters are the most widely used detectors for thermal neutrons. However, ^3He is an extremely rare isotope of helium with a natural abundance of about 1.37 ppm. Due to the dwindling supply of ^3He , now there is an urgent need to explore alternative materials [2,3]. The other gas detector used for thermal neutrons is based on BF_3 [4]. However, the corrosive and toxic nature of BF_3 is a problem with this kind of detectors. Thus alternate neutron detectors based on plastic or liquid organic scintillators, inorganic, glass or other scintillator materials are being explored for applications in neutron detection [5–9].

Thermal neutron detectors with compact size and high efficiency are desirable. In this regard inorganic scintillator single crystals are promising candidates to fabricate portable and durable solid state detectors [10,11]. Apart from this, the higher atomic density as compared to conventional gas-filled detectors may result in higher detection efficiencies even with smaller sizes. Suitably engineered materials can also help to impart insensitivity to gamma radiation, which is a potential problem with solid state devices

compared to gas-based detectors. Single crystals of $\text{Li}_6\text{R}(\text{BO}_3)_3$ ($\text{R} = \text{Gd}, \text{Y}$ and Lu) doped with cerium have been proven as a promising neutron scintillator [12]. This material contains ^6Li (natural abundance: 7.4%; $\sigma_{\text{abs}}: 940 \text{ b}$) and ^{10}B (natural abundance: 20%; $\sigma_{\text{abs}}: 3835 \text{ b}$) that have large cross-sections for thermal neutrons and produce charge particles (^4He and ^3H) after neutron interaction [13]. The charged particles generated in ^{10}B (n, α) ^7Li and ^6Li (n, α) ^3H reactions excite the Ce^{3+} ions resulting in a fast and efficient emission at about 415 nm. The efficiency response of bi-alkali photomultiplier tubes (PMT) matches well at this emission and therefore can be gainfully employed as a read-out device using standard electronics. Among them $\text{Li}_6\text{Gd}(\text{BO}_3)_3$ has the maximum light output which is around five times that of $\text{Li}_6\text{Y}(\text{BO}_3)_3$. However, the emission of gamma cascade (up to 8 MeV) and conversion electrons due to the interaction of ^{157}Gd with low energy neutrons is main disadvantage of these materials [14]. The single crystal of LYBO:Ce has a reasonable light output of about 1200 photons/MeV and replacement of “Gd” by “Y” having a lower capture cross-section for neutrons enables a clean pulse height spectrum [13]. A lower effective atomic number (Z_{eff}) of the LYBO as compared to other solid state devices also helps to impart a lower sensitivity to gamma-rays that commonly exists along with neutrons [14]. Most of the measurements reported for these crystals are carried out with gamma and alpha sources to show the feasibility of neutron detection. However, actual detection of neutrons using the scintillator crystals has not been demonstrated in most of the reported literature [15]. In the present work we report the growth of good quality single crystals of $\text{Li}_6\text{Y}(\text{BO}_3)_3\text{:Ce}$ and fabrication of compact

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detectors to detect thermal neutrons over a wide range of flux from about $10 \text{ n/cm}^2/\text{s}$ to $10^7 \text{ n/cm}^2/\text{s}$.

2. Experimental details

Cerium doped (0.1 mol%) single crystals of LYBO were grown using the Czochralski technique in an automatic diameter controlled crystal puller system (Model: Oxypuller, Cyberstar) [16]. Synthesis of the starting charge in the form of a single phase material was carried out from high purity constituents Li_2CO_3 , Y_2O_3 , H_3BO_3 and CeO_2 taken in a stoichiometric ratio and using the solid state sintering process. The materials were taken in their natural abundance of Li and B. A two step sintering at 700°C for 24 h each was used with intermediate mixing to ensure the formation of the single phase material [17,18]. The phase and the thermal behavior of the sintered charge and the as-grown crystal were verified using the Differential Thermal Analyser (SETRAM make DTA-TG 92). The melting and solidification behavior were recorded at a heating/cooling rate of 10 K/m in N_2 ambient. The phase purity of the synthesized material and the grown crystal was verified using powder XRD data, employing the Rigaku RINT 2000 diffractometer (Cu $\text{K}\alpha$ radiation) with a step size of 0.02° in 2θ range of 10 – 80° . The growth is carried out in a platinum crucible under argon ambient using an LYBO crystal seed oriented along $(1,0,\bar{2})$. A RF power supply was used to melt the material by induction heating. The melt temperature was kept approximately 50°C higher than the melting point to homogenize the melt which is recommended for the growth of borates crystals [13].

The grown crystals were characterized by measuring the transmission and absorption spectra of a 2 mm thick disk, cut and polished from the crystal, using a Shimadzu 3600 UV–vis NIR spectrometer in the range from 185 nm to 800 nm. Photoluminescence (PL) studies were performed over a wavelength range from 250 nm to 800 nm at room temperature employing a fluorescence spectrometer (Edinburg Model-FLP920). The emission was recorded by positioning the sample at 45° with respect to the excitation beam. A steady state xenon lamp was used as an excitation source and a spectral bandwidth of 1 nm was selected for both excitation and emission arms. The recorded luminescence spectra were corrected for the spectral sensitivity function of the instrument. A nitrogen gas filled flash lamp operating at a repetition rate of 40 kHz was employed for recording the life-time characteristics of the photoluminescence decay. The samples have been excited at a wavelength of 360 nm using nitrogen emission lines and the temporal response of the luminescence at 410 nm was recorded. The instrument response function was also recorded by keeping a reflector in place of the sample.

To measure the scintillation characteristics, samples of 20 mm diameter and 2 mm thick were processed from the as-grown crystal ingot. A thinner sample was taken to minimize the interaction with gamma radiation. However, higher atomic density of lithium and boron atoms in these materials ensured excellent efficiencies for the interaction with thermal neutrons. One face of the crystal samples was polished and optically coupled to a photo-multiplier tube (Hamamatsu; Model no. R2154). The scintillator crystal was wrapped with multiple layers of reflecting tapes and was finally wrapped with an aluminum foil. The reflecting tapes ensured an efficient collection of light towards the PMT while Al foil minimizes the PMT dark current due to the background. A pulse processing chain comprising a pre-amplifier, a shaping amplifier and a 8k multi-channel analyzer (MCA) was used to measure the pulse height spectra (PHS). The PHS was recorded for neutrons from the Dhruva nuclear reactor with a flux of about 10^7 n/s/cm^2 . Radiation shields like lead brick (for gamma-rays) and borated rubber (for neutrons) were used to discriminate spectra originating from neutron and gamma-rays, respectively. The pulse height spectra have also been recorded for

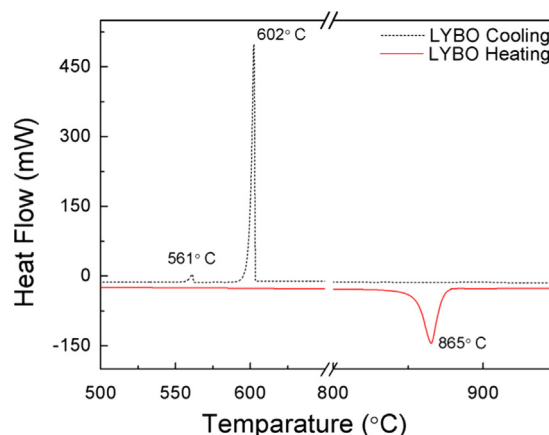


Fig. 1. DTA plots of LYBO:Ce material (solid line) heating and (dashed line) cooling cycle.

other neutron sources such as ^{252}Cf and Am–Be that have lower flux values. The scintillation decay of the LYBO:Ce crystal was determined by recording the anode pulse of the PMT using a digital storage oscilloscope having 1 GHz bandwidth (Tektronix, MDO-3102). The decay time was measured for both neutrons and gamma exposures.

3. Results and discussion

3.1. Crystal growth

The melting and the freezing behavior of the LYBO:Ce is shown in Fig. 1. The DTA plot shows a large difference of about 250 K in the melting (865°C) and the freezing peak (602°C). This suggests that a large super cooled liquid is required to initiate the crystal growth process.

A photograph of an as-grown single crystal of LYBO:Ce is shown in Fig. 2. The grown crystals have diameter of about 20 mm and 25 mm length. The crystals are free from any visible inclusion or impurity. The optimum growth was achieved with a pull rate of 0.5 mm/h and a rotation of 10 – 20 rpm . Due to high viscosity of the melt, the formation of bubbles is a common problem in borate single crystals. The bubble entrapment in the growing crystal has been effectively eliminated by using a high temperature gradient (about 100°C/cm), a slower pull rate and a relatively higher rotation rate [19–20].

The presence of cleavage planes along $(1,0,\bar{2})$, $(0,1,0)$ and $(1,2,1)$ directions increases the tendency of the crystal to develop cracks either during the growth/cooling or while processing [20]. Therefore a seed oriented along $(1,0,\bar{2})$ was used to avoid the cleavage along the length of the crystal. To minimize thermal stresses produced during the growth and reduce cracking during processing the as-grown crystals ingots were annealed at 500°C for 24 h [13].

3.2. Optical characterization

For the measurement of scintillation properties the crystal ingot was cut into discs of 20 mm diameter and 2 mm thickness and polished to optical finish using alumina powder abrasive. The transmission, emission and excitation characteristics of an LYBO:Ce crystal are shown in Fig. 3. The transmission for a polished sample with a thickness of 2 mm was more than 80% in the wavelength range from 185 nm to 1100 nm (shown only up to 600 nm in the figure). The emission peak at around 415 nm is

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