ABSTRACT

Crystals are the pillars of modern technology; crystal growth is a fascinating field of research. The major advancement in scintillation detection came with the development of good scintillation crystals and efficient photomultipliers. One of the primary challenges in the field of detectors is to select a material with higher scintillation efficiency and fast decay time characteristics. Many materials already exist with good characteristics. Among them organic scintillators have good scintillation efficiency with fast decay time. Materials like anthracene and stilbene are being used in the field of scintillation detectors for the past few decades. These crystal scintillator instruments have opened many areas of particle nuclear physics and high-energy physics. Due to low effective atomic number, organic materials in contrast to inorganic ones are characterized by negligible back scattering effect for the process of charged particles absorption. High anisotropic nature and directional recoils of these materials has led to the detection of different types of particles in the various radiation backgrounds. Also, this property has led to the detection of various light particles to weakly interactive massive particles (WIMPS) from the dark matter halo. Due to high concentration of hydrogen atoms they are used in fast neutron, beta, alpha spectroscopy. But still the commercially available crystals have many defects and the property variation with regard to quality of grown crystal was not studied extensively. So it is of specific interest to study the property of the crystal by adopting different suitable growth techniques.

Organic scintillators have several important properties that make them particularly useful for wide range of applications. The fast scintillation decay time of the order of few nanoseconds, enables them to be used in conjunction with fast photomultipliers to provide scintillation counters with time resolution less than nanoseconds. Linear response to electrons, high hydrogen content, dependence of the pulse shape on stopping power, the emission spectra matching well with the S11 photocathodes, the low density, low atomic number and reduced scintillation efficiency to heavy particles are specific properties of organic crystal scintillators.

The present investigation deals with the growth of organic scintillation crystals and their characterization. The application of the crystals in particle detection has been established through experiments. Electron momentum distribution of the grown crystal has been established. The whole of the work has been divided into seven chapters. Here we present the conventional growth methods adopted for the growth of organic molecular crystals, anthracene and stilbene with preliminary characterizations of the grown crystal. Attempts have been made with modified methods for the growth of stilbene and anthracene and the grown crystals are characterised. X-ray rocking, SEM, Optical microscope, Raman spectroscopy and photoluminescence studies have been carried out and the results obtained are discussed. Scintillation detector experiments were done for the grown crystals using high-energy proton beam, the energy response and rise time response were carried out. Electron momentum distribution of the grown crystal has been mapped at room temperature, 210 K and 110 K.

Trans-stilbene crystals were grown by vertical Bridgman technique (VBT). The successful growth depends on the purity of *trans*-stilbene, shape of growth vessel and growth rate. The progressive lowering of growth rate ranging from 2 to 0.5 mm/h yields single crystals. The ampoule with a cone angle varying from 20 to 25 ° and with bent capillary inclined at an angle of 23 to 27 ° produced reproducible yield of single crystals. The crystals are in the form of boules 5 cm long and 2 cm diameter and could be cleaved easily along (001) plane. FTIR spectrum was recorded to identify the functional groups present in the stilbene crystals. The optical transmission spectra show a transmission efficiency of 57%. X-ray diffraction studies were carried out for the grown ingot and the lattice parameters were calculated. From the timing resolution studies it was confirmed that the crystal element has the timing resolution of 7.5 ns.

Good transparent crystals of *trans*-stilbene and anthracene were grown using modified VBT. The modifications in the length of the furnace were helpful in the yield of lengthy crystals free of defects. The continuous decrease in the ampoule translation rate was helpful in sustaining flat interface through out the growth and it helps in the yield of highly transparent crystals. Good seeding was done using the double walled crucible with additional necking facility. The grown crystals were characterized using powder X-ray diffraction studies NMR studies, and uv-visible transmission studies. Improved crystal growth methodology has brought out the improvement in the timing characteristics of the phosphor.

A selective self-seeding technique in VBT (SSVBT) was proposed and adopted for the growth of *trans*-stilbene crystal. The double walled ampoule with inner tube having slightly bent and necked capillary tube with its tip having a distance more than 7mm from outer tube tip always yields single crystal. Multiple twinning, multi nucleations, impurity effects on nucleation were all sorted out by the above-proposed method. Seed selection and necking to reduce defects were also done automatically by this method. The double crystal X-ray diffractometry (DCD) results exhibit low FWHM value of 32.3 arcsec for the crystals grown by the present method and the crystal grown by normal conventional method shows FWHM value of 58.7 arcsec. This DCD result confirms that the crystal grown by this novel method is of better quality than the crystal grown by conventional technique. The time resolution of selective self seeded crystal for gamma ray of 511 keV is better than that of conventional method grown crystal.

Double Run Selective Self Seeding Vertical Bridgman Technique (DRSSVBT) has been proposed and it was established for the growth of anthracene. The quality of the grown crystal was established by time resolution studies. The etching studies exhibit rarefied etch pits and there are no low angle grain boundaries. Quality assessment of crystals grown by DRSSVBT was made using the double crystal X-ray diffractometry and timing resolution studies. The rocking curve of the conventional method grown crystals exhibits FWHM of 46.3 arcsecs. For the DRSSVBT grown crystals X-ray rocking curve exhibits FWHM of 29 arcsecs. Additional low intense peak in the rocking curve indicates the low defects due to effective control of thermal defects in DRSSVBT grown crystal.

The micro hardness anisotropy and plastic deformation have been observed on Vertical Bridgman grown *trans*-stilbene single crystals using Vicker's micro hardness studies. It has been observed that plastic deformation occurs by means of translational slip. The anisotropy of planes is observed by the different slip systems that are different in different planes. The systems are $(2\ 0\ \bar{1})$ [010] and (100) [010]. The results obtained on the stilbene single crystals are in accordance with observations on the other molecular solids that possess same structure and space group. The angles observed between different slip planes and directions prove the slip traces.

The vertical Bridgman grown *trans*-stilbene crystal quality has been analyzed with respect to its growth aspects. The quality of the grown crystal has been studied and it was found that slow seeded crystals with full conical shaped ampoules have uniform quality throughout. Orientation changes in the crystals take place due to the change in the seeded region of the capillary. The slow seeded crystal grown with conical ampoule exhibits rocking curve of FWHM 27 arcsecs. Optical microscopic study shows very rare defects through out the crystal. Poor quality crystals were obtained with faster growth rate and ampoule design different from the optimized one. The studies bring out that the crystal grown with ampoules fully conical with twisted seeding from capillary (selective self seeded) and gradual decrease of growth rate from 2.0 - 0.5 mm is uniformly good quality crystal. But the top region of the crystal of about 1.0 - 0.5 cm exhibits X-ray rocking curve width of 60-70 arcsecs. The higher FWHM may be due to fast supercooling in the upper open surface, fast quenching and little trace of low-density impurities. Raman phonon studies have been carried out to identify the shift in phonon modes due to irradiation. The results show that the grown crystals are having more ordered structure when compared to previous reports. Defect induced modes were observed for proton irradiated and gamma irradiated crystals. Intensity reversal for molecular modes due to irradiation has been observed. In solution grown anthracene crystals, defect luminescence background is very high and it depletes the Raman modes. Defect luminescence background gets reduced with improvement of growth method. It gets reduced by more than 25 times in vapour grown crystals when compared with solution grown crystals. It gets reduced by 3 times in conventional Bridgman technique grown crystal when compared with the vapour grown crystal and it is absent in DRSSVBT grown crystal. The Raman modes corresponding to lattice and molecule become more distinct with decrease in the defect luminescence.

High performance, device quality stilbene single crystals were obtained from the selective self-seeding technique. The quality assessment made by timing resolution gives the timing resolution of about 2.23 ns, which is an improvement from about 7 ns for the crystal grown by conventional method. The decay time calculated from the timing spectrum is found to be 1.6 ns. This is the lowest decay time reported for gamma scintillation for stilbene. The good n-γ and α- γ discrimination has been obtained for ^{252}Cf and ^{241}Am sources.

Scintillation response to high-energy protons has been done. It was observed that nearly linear response has been seen for high-energy protons, 5 to 25 MeV. Rise time spectroscopy of trans-stilbene to high-energy proton using commercial pulse shape discrimination (PSD) module has been done. The experimentally observed rise time shift does not coincide with the theoretical prediction given by Papadopoulos. The rise time shift for different proton energy ranging from 5 to 25 MeV in stilbene has been done for the first time. It was observed that the rise time decreases with increase in the energy of proton. The results are contradictory to the theoretical prediction found already in reports.

Positron two-dimensional angular correlation studies have been carried out and the electron momentum distribution of the grown *trans*-stilbene crystal with c-a plane projection has been extracted for the first time at room temperature, 210 K and 110 K.