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Growth and characterization of benzyl 4-hydroxybenzoate single crystal by vertical Bridgman technique for optical applications

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ABSTRACT

The benzyl 4-hydroxybenzoate single crystal has been grown by vertical Bridgman technique. The grown crystal was confirmed by single crystal X-ray diffraction studies. The presence of functional groups in the crystal was confirmed by Fourier transform infrared (FTIR) spectral studies. The thermal behaviour of the grown crystal was analyzed by thermogravimetric analysis (TGA), differential thermal analysis (DTA) and differential scanning calorimetric (DSC) studies. Optical behaviour of the grown benzyl 4-hydroxybenzoate crystal was studied by UV–Vis-NIR spectral analysis. Fluorescence spectrum shows near violet light emission. The second harmonic generation behaviour of benzyl 4-hydroxybenzoate was analyzed. The laser damage threshold value of benzyl 4-hydroxybenzoate was measured as 2.16 GW/cm^2. The dielectric measurements of benzyl 4-hydroxybenzoate crystal were carried out with different frequencies 1 kHz to 1 MHz versus different temperatures ranging from 313 to 353 K. Photoconductivity study shows that the grown benzyl 4-hydroxybenzoate crystal belongs to negative photoconductivity property. The mechanical strength of the crystal was calculated by Vickers microhardness study.

1. Introduction

In recent years great efforts have been made to explore the molecules with excellent nonlinear optical (NLO) properties due to their widespread applications such as frequency conversion, telecommunications, optical logic gates, laser radiation protection, frequency mixing, optical parametric oscillations and optical data storage, etc., [1,2]. Organic NLO materials are attracting a great deal of attention because of their flexibility in molecular design, high nonlinearities and rapid response in the electro optic effect compared to inorganic [3]. Nowadays researchers are searching new NLO crystals for laser applications [4]. The benzoate family materials are excellent nonlinear optical properties such as methyl p-hydroxybenzoate [5], 2-amino 4-picolinium 4-aminobenzoate [6], ethyl p-amino benzoate [7], piperidium p-hydroxybenzoate [8], ethyl p-aminobenzoate [9], 2-amino pyridium 2-chloro 4-nitrobenzoate [10], benzimidazolium p-hydroxybenzoate [11], and butyl 4-hydroxybenzoate [12]. In this series we have choose benzoate family material benzyl 4-hydroxybenzoate. Benzyl 4-hydroxybenzoate is a prominent material which is non-hygroscopic, its molecular formula ($C_{14}H_{12}O_3$), its molecular weight (228. 24 g/mol) and it has non-centrosymmetric structure with space group $P2_12_12_1$. The benzyl 4-hydroxybenzoate possesses carbonyl group (C═O), in this group emits fluorescence properties [13]. The compound possess 820 isomer structural molecules [14], such as 2-(o-Tolyloxy)benzoic acid [15], trans-Resveratrol [16], 2-(Biphenyl-4-yloxy)acetic acid [17], Xanthyletin [18], 2′,2′-dimethyl-3-pyreno[6,5:7,8]coumarin [19], (E)-3-(Furan-2-yl)-1-(4-methoxyphenyl)-prop-2-en-1-one [20], 2-hydroxy-4-methoxybenzophenone [21,22], etc., were its molecular formula, molecular weight are same, but different structural formula.

Different crystal growth techniques are used to grow organic single crystals. For device fabrication large size crystals are needed. The solution growth is most common for organic nonlinear optical single crystals but some crystals obtained small size and affect solvent inclusion problems. However, growing bulk defect free single crystals of organic material from the melt growth methods is also a challenging task due to its low thermal conductivity. Hence, for the bulk organic single crystal growth, melt growth by vertical Bridgman processes are most preferable. In vertical Bridgman technique ampoule designs and translation rates are optimise to grow large size good quality organic single crystals [23]. The growth technique is decided mainly depending upon the physical and chemical properties of the materials. From the TG-DTA and DSC spectra shows that there is no weight loss occurs before the melting point.

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So the benzyl 4-hydroxybenzoate was decided to grow vertical Bridgman technique. In this present work the organic material benzyl 4-hydroxybenzoate was grown by vertical Bridgman technique and the grown crystal was subjected to various characterization techniques such as single crystal XRD, FTIR, UV–vis-NIR, fluorescence, SHG, laser damage threshold, photoconductivity, dielectric, and Vickers micro hardness.

2. Experimental

The organic material benzyl 4-hydroxybenzoate was successfully grown by vertical Bridgman technique. In vertical Bridgman technique, the ampoule design is one of the important parameter, the different types of ampoules are designed [24–27] and grown single crystals. A borosil glass was used to design the ampoule. The ampoule cone length is 4.5 cm, above the cone is 19 cm height with an inner diameter of ampoule is 1.5 cm.

The organic material benzyl 4-hydroxybenzoate (380709 Aldrich) was filled into the ampoule and it was vacuumed to 10^{-4} torr using a rotary vane pump and sealed. The furnace temperature was maintained above the melting point of the benzyl 4-hydroxybenzoate material for 24 h due to in order to neglect the bubble formation during the growth period. The material reach the homogeneous melt state then translation is started. The ampoule was moved from hot zone to the cold zone in mm level using nano translation assembly. We tried three translation rates such as 0.6 mm/h, 0.5 mm/h and 0.4 mm/h and optimized this material as the translation rate 0.4 mm/h got the good crystal. The growth was directly observed through the furnace. After the completion of growth, the furnace temperature was reduced to room temperature with the cooling rate of 1 °C/hr because it helps to avoid the cracks in the grown crystal due to the difference in the thermal expansion coefficient between the ampoule and crystal. The crystal of size 80 mm length and 15 mm diameter has been successfully grown. The cut and polished benzyl 4-hydroxybenzoate crystal is shown in Fig. 1.

3. Results and discussion

3.1. Single crystal X-ray diffraction studies

The grown benzyl 4-hydroxybenzoate crystal was subjected to the Single crystal X-ray diffraction studies. The single crystal X-ray diffraction data were collected using Enraf Nonius – CAD4 single crystal X-ray diffractometer. The observed lattice parameters of the grown crystal were good agreement with the literature [28]. The grown benzyl 4-hydroxybenzoate crystal belongs to orthorhombic system with the non-centrosymmetric space group $P2_12_12_1$. The obtained values are shown in Table 1.

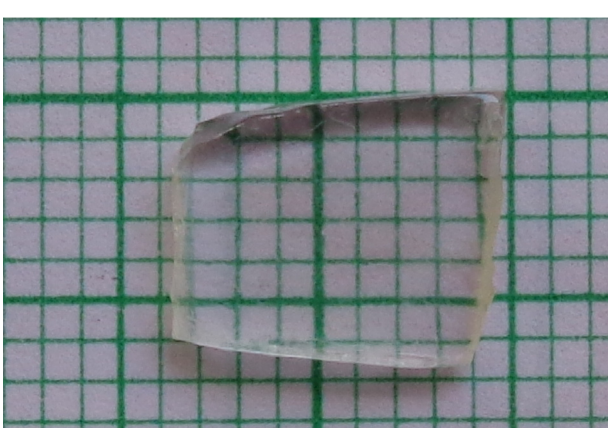

Fig. 1. Cut and polished portion of the grown benzyl 4-hydroxybenzoate single crystal.

Table 1
Single crystal XRD data for grown benzyl 4-hydroxybenzoate.

Parameters	Literature [11]	Present value
a (Å)	8.514 (1)	8.55 (±0.010)
b (Å)	22.753 (3)	22.72 (±0.03)
c (Å)	5.874 (1)	5.87 (±0.006)
α (°)	–	90
β (°)	–	90
γ (°)	–	90
Volume (Å ^{3})	1137.9 (4)	1140 (±4)
Crystal system	Orthorhombic	Orthorhombic
Space Group	$P2_12_12_1$	$P2_12_12_1$
Number of molecules per unit cell	4	4
Density (mg m^{-3})	1.332	1.329

3.2. FTIR spectral studies

Fourier Transform Infrared Spectroscopy (FTIR) was used to analyze the functional groups of the grown benzyl 4-hydroxybenzoate crystal for the range of 4000–400 cm^{-1} by Perkin Elmer spectrometer from the KBr pellet technique. Fourier Transform Infrared Spectroscopy (FTIR) spectrum of benzyl 4-hydroxybenzoate was shown in Fig. 2. The peak obtained at 3383 cm^{-1} is due to O—H stretching vibration. The strong peak observed at 1684 cm^{-1} is due to C═O stretching of ester. The peak obtained at 1587 cm^{-1} is due to C═C stretching. The peak observed at 1441 cm^{-1} is due to H—C—H stretching. The peak observed at 1208 cm^{-1} is due to C—O—O asymmetric stretch of phenol esters and peak at 1157 cm^{-1} is due to O—C—C asymmetric stretch of benzyl esters. The peaks at 852 cm^{-1}, 770 cm^{-1} and 697 cm^{-1} are due to aromatic C-H stretching [29,30].

3.3. Thermal analysis

The thermal behaviour of the grown crystal was analyzed by using TG/DTA and DSC analyzes. The Q600 SDT thermal analyzer is used to analyze the thermo gravimetric (TG) and differential thermal analyzes (DTA). The sample was measured at the temperature between 28 °C and 350 °C. The results obtained from TG/DTA are shown in Fig. 3a. The TG analysis shows single stage weight loss starts around 177.35 °C and below this temperature no significant weight loss is observed. The DTA reveals that there is sharp endothermic peak observed near 114 °C was assigned to the melting point of the grown crystal. The second endothermic peak observed around 299 °C was assigned as the decomposition point, in this stage heavy weight loss in TG was observed. From DTA observed that there is no exothermic or endothermic peak up to the melting point. It was observed that there is an absence of phase transition or decomposition up to the melting point. Differential scanning calorimeter (DSC) analysis was carried out by using Q20 thermal analyzer. The observed DSC curve is shown in Fig. 3b and the results shows that the endothermic peak (heating curve) around 114 °C which is the melting point of the material and the exothermic peak (cooling curve) around 56 °C which is the crystallization point of the benzyl 4-hydroxybenzoate. The super cooling temperature is calculated using the standard formula:

$$\Delta T = T_m - T_c \tag{1}$$

where ΔT is the super cooling temperature, T_m is the melting point and T_c is the crystallization point.

$$\Delta T = 114^\circ C - 56^\circ C = 58^\circ C \tag{2}$$