ISSN: 0974-5823 Vol. 7 No. 4 April, 2022

International Journal of Mechanical Engineering

SECOND AND THIRD ORDER NONLINEARITY OF ETHYL P-AMINOBENZOATE (EPAB) SINGLE CRYSTAL GROWN BY SOLUTION AND BRIDGMAN TECHNIQUE – A COMPARATIVE ANALYSIS

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ABSTRACT

By using vertical Bridgman technique and slow evaporation solution growth technique high quality single crystals of ethyl p- amino benzoate has been grown. The second order and third order nonlinear optical behavior has been compared. The crystals grown by solution growth technique has shown a higher value of second order and third order nonlinear optical behavior.

Keywords: Single crystal; Z-Scan; NLO;

1. INTRODUCTION:

NLO materials have fascinated and are gaining enormous demand due to their vast applications like optoelectronics, laser technology, and data storage [1]. In both past and current years NLO is one of the most interesting and attractive fields in the research because of its molecular flexibility, nonlinear optical structure, and optical computing. Organic materials have shown significant promise due to their vast-ranging properties than those of inorganic counterparts.[2] The organic and inorganic materials are differing in their hyperpolarizability values. Therefore organic materials have a high hyperpolarizability value compared to their inorganic counterparts because of their orientation and delocalization of π electron in nonlinear optics [3]. It has large applications such as rapid response time, optical control, optical transmission, etc. Third Harmonic Generation (THG) is a completely electronic and coherent process. Since all materials have non vanishing third-order susceptibilities. It captures the lead of NLO like carbon-based materials, liquid crystal, dielectric and organic dye. And it is used to study the solely electronic molecular second hyperpolarizability of centrosymmetric materials. It is used to evaluate the magnitude and indication of real and imaginary parts of nonlinear optics [4].

Over the past few years, ethyl p- aminobenzoate (EPAB) was grant as a successive organic NLO material. It has an orthorhombic crystal system with the molecular formula $C_9H_{11}N_2$. [5,6]. Solution growth is an adaptable technique that allows the growth of the high aspect single crystal. Arivanandhan et al. made the first single crystal by examining the czochralski technique. Bridgman growth is an affinity method for the composing of large single growth of an extensive variety of substances including the organic crystal. AnandhaBabu et al. and Muthuraja et al. have done a study on the organic crystal EPAB (Ethyl p- aminobenzoate) by the Bridgman technique [7]. However to our knowledge no reports are available for the comparison study of vertical Bridgman and solution growth using Third Harmonic Generation (THG) of ethyl p – amino benzoate single organic crystal. In the present paper, we proposed a comparisons study between the vertical Bridgman technique (VBT) and the Slow Evaporation solution growth technique (SEST) of Ethyl p – amino benzoate (EPAB) single crystal.

2. EXPERIMENTAL:

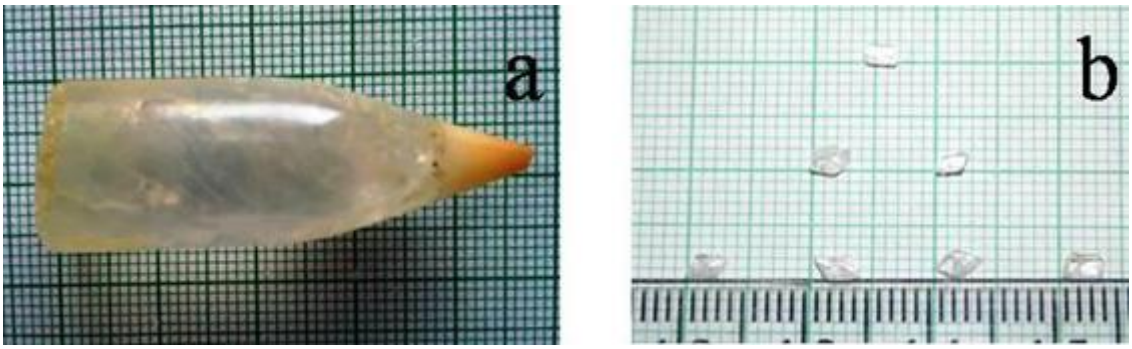

Fig 1: (a) EPAB grown crystal by Bridgman (b) EPAB grown crystal by solution growth

The Bridgman growth EPAB materials are commercially brought from Sigma Aldrich. The sharp cone-shaped ampoule was used and source materials were filled in the borosil ampoule. The material–packed ampoule was evacuated in the range of 10^{-6}torr and sealed. Approximately 10^0 to 20^0 C temperatures were maintained inside the two-zone boiler and it is higher than the substance melting point. Inside the furnace, the sealed ampoule was hanged in the hot region. Due to the thermal conductivity and slow growth rate the organic single crystal is grown using the Bridgman technique.

The translation rate is less than 1.2 mm/hour. Once the crystal is grown the furnace were cooled to room temperature. From the ampoule, the crystal was carefully removed by using a standard diamond wheel cutter. And it took around 22 days to grow as shown in Fig. 1the slow evaporation solution growth EPAB was commercially available from E-MERCK India. Ltd. For the solution method selection of solvent plays an important role in the growth of the organic crystal. At the present investigation, ethanol is used as a solvent for growing EPAB organic single crystal. It took around 26 days to grow a single crystal as shown in the figure. 1 (a) and (b)

3. Z-scan measurements

Z-scan technique is an easy and efficient technique compared to other analyzes [8]. This is standard techniques to determine various parameters such as non-linear absorption coefficient, third order nonlinear refractive index for solid crystals. The z scan technique is the exact method to find out both non linear absorption coefficient (β) and non linear refractive index (n_2). The grown crystals were analyzed through He-Ne laser of wavelength 632.8 nm. The input laser modifier to Gaussian form using Gaussian filter [9]. Through the open aperture and close aperture were measured third order nonlinear susceptibility.

The testing sample was transferred by using stepper motor from +Z to – Z axis. The nonlinear absorption and nonlinear refractive index can be measured from the close and open aperture z scan technique. The third-order nonlinear refractive index and nonlinear absorption coefficient of SEST and VBT grown single crystal were determined by z scan technique using Nd-YAG LASER at 532nm.

The open aperture analysis is used to estimate the non-linear absorption coefficient. The Open aperture curve shows Fig. 2. (a and b). The refracted laser beam is entirely absorbed by the detector in the open aperture method. Fig 2. Shows analyze the intensity of transmittance was found very high at the focal point {Z= 0}. The minimum transmittance (valley) specifies Multiphoton absorption and it shows intensity-dependent absorption which is due to either Multiphoton absorption. Fig 2. b curve shows maximum transmittance (peak) and it shows the saturation absorption. The transmitted light was measured by the detector where it is sensitive only to the intensity variation. From the above figure the flat response for a purely refractive non-linearity is detected by open aperture.

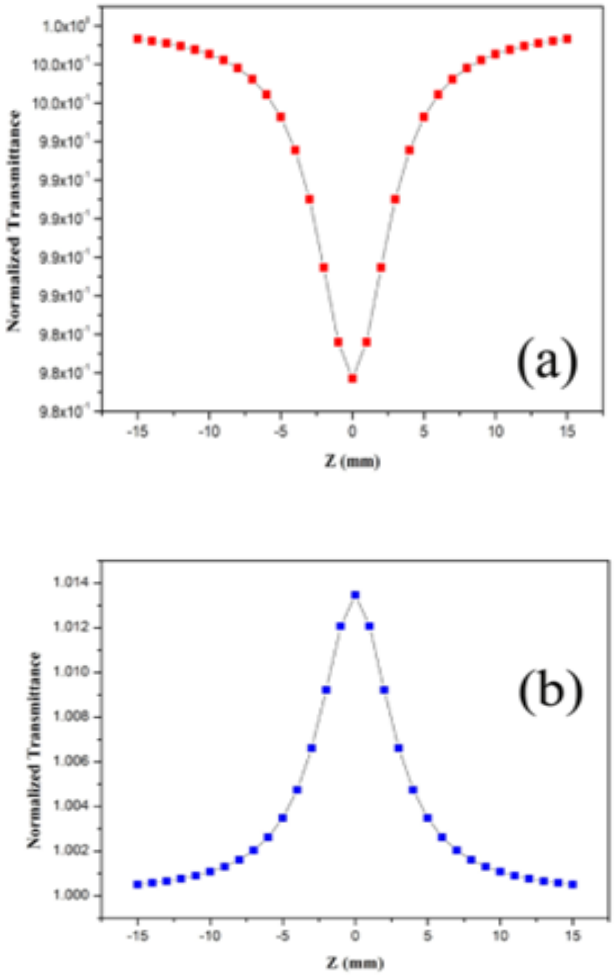

Fig. 2 (a) open aperture of Solution growth, (b) open aperture of VBT.

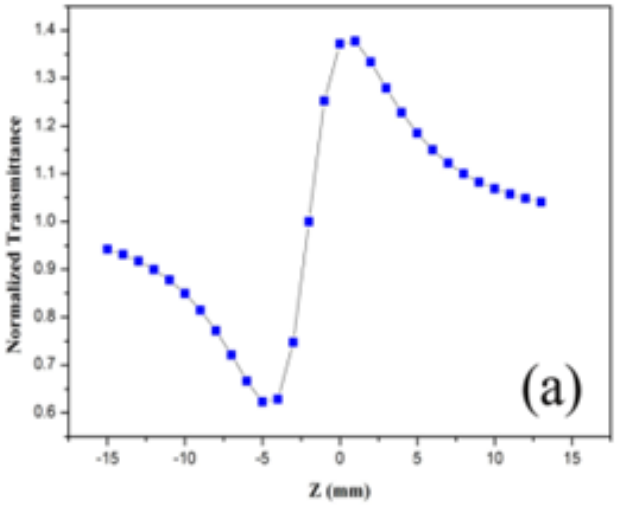

Fig.3 (a) Self-focusing effect of SEST crystal in closed aperture,

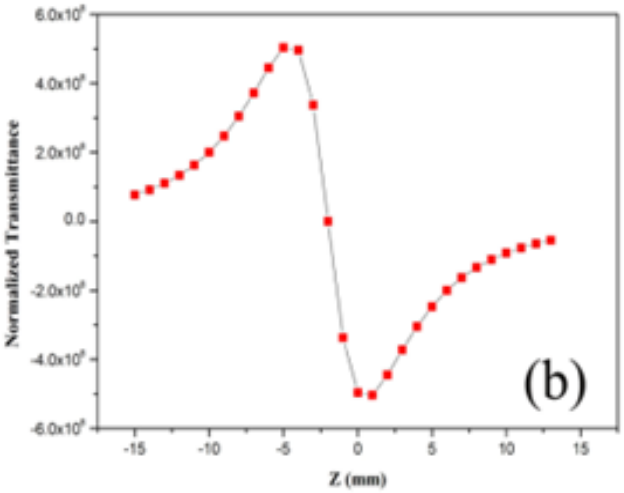

Fig.3 (b) Self-defocusing effect of VBT crystal in closed aperture.

The closed aperture analysis gives the characteristic nature of the non-linear refractive index of the sample. The closed aperture curve shows the valley followed by peak *configuration* and it is shown in Fig 3. a. The Valley followed by peak configuration reveals the self-focusing effect. The closed aperture, analyze indicates the asymmetric peak which is also known as the self defocusing effect and it is shown in Fig 3. b. The negative non-linear refraction of the sample self defocusing effect has been elucidated. The local variation of the refractive index with temperature attributes to the self-defocusing effect [10]. Due to the self defocusing effects, the grown crystal is more suitable for optical sensors and night vision devices. [11]

The experimental details and results of the Z-scan technique for SEST and VBT grown EPAB crystal are given in Table 1. From the above analysis, it was concluded that the EPAB crystal grown from VBT is having a high value of third order nonlinear susceptibility when compared to the SEST grown EPAB crystal.

Table 1 Comparative study analysis for VBT & SEST

Growth method	**Third order nonlinear susceptibility (χ^3)**	**Nonlinear refractive index (n_2)**	**Nonlinear Absorption coefficient (β)**	**Growth period (Days)**
VBT	7.33421X 10^{-6}esu	1.01x10^{-11}m/w	5.97 X 10^{-5} m/W	22
SEST	1.00678x 10^{-5}esu	1.43x10-^{11}m/w	8.11 X 10^{-5} m/W	26

From the above table, we concluded that the EPAB Crystal grown from SEST (1.00678X 10^{-5}esu) having high Third order nonlinear susceptibility when compared to the VBT (7.33421X 10^{-6}esu) grown EPAB crystal.

4.Second Harmonic Generation

The SHG efficiency of the grown EPAB crystal (VBT and SEST) is measured with respect to KDP by using ND-YAG laser (1064nm). The input of laser energy is 0.68J and the SHG values for crystals grown by VBT is 7.96 which is greater than KDP value (7.80). The input laser energy value for SEST grown crystal is 0.87J. The SHG value is 8.01 which is greater than KDP (7.80) and VBT crystals.

5. CONCLUSION:

Organic NLO material Ethyl -p amino Benzoate (EPAB) with molecular formula $C_9H_{11}N_2$ were grown by the solution growth technique and Bridgman technique. We have shown the comparison study of vertical Bridgman growth technique and slow evaporation solution growth technique. The experimental details and results of the Z-scan technique for SEST and VBT grown organic single crystal were analyzed

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