Optical and piezoelectric studies of an efficient frequency doubler: Imidazolium L-Tartrate (IMLT).

A. Jagadesan[a], R. Subhashini[a], Santhi M George[a], G. Peramaiyan[b], S. Arjunan[c]

[a]*Department of Physics, R.M.K Engineering College, Kavaraipettai, Tamil Nadu - 601206*
[b] *Postdoctoral researcher, Institute of Physics, Academia Sinica, Taiwan.*
[c] *Department of Physics, TJS Engineering College, Peruvoyal, Tamil Nadu - 601206.*
Corresponding Author E-mail : ajn.sh@rmkec.ac.in

Abstract

Good quality crystal of imidazolium L-tartrate (IMLT) with a large size of 34×25×22 mm^3 has been grown by the slow cooling cum seed rotation solution growth technique. The morphology of IMLT crystal has been indexed by single crystal X-ray diffraction study which revealed the monoclinic crystal system with major forms of (0-10) and (001). Phase matching angle of IMLT was found to be 42±0.2° using a Nd:YAG laser and it was also found that the IMLT crystal belongs to type-1 phase matching category. From the piezo-electric study, the direct (d_{33}) and transverse (d_{31}) charge coefficients were calculated to be 1 pC/N and 17 pC/N, respectively.

Keywords: Crystal growth, Nonlinear optical material, Piezoelectric materials

1. Introduction

Nonlinear optical (NLO) crystals are playing an important role in the field of laser science and technology due to their potential applications such as second harmonic generation (SHG), third harmonic generation (THG), fourth harmonic generation, difference frequency generation (DFG), etc [1-3]. Some series of typical inorganic and semi-organic NLO crystals, such as potassium dihydrogen phosphate (KDP), potassium pentaborate (KB_5), β-BaB_2O_4 (BBO) and L-arginine phosphate monohydrate (LAP) have been used for second harmonic and UV generations due to their high transparency in UV-Vis region, moderate laser damage threshold, thermal stability and phase matchability [4-6]. Even though inorganic crystals have many preferred properties, they have been limited in device fabrications due to low second and third harmonic generation efficiencies. Generally, organic acentric molecular materials have large hyperpolarizability and bulk nonlinear optical susceptibility due to the existence of π-conjugated system with a donor and an acceptor group at the two ends of the conjugation [7-9]. But they are also limited to device fabrications due to their poor thermal and mechanical stabilities, etc. But some series of L-tartrate based organic crystals have been found to have good thermal and mechanical stabilities. In this series imidazolium tartrate (IMLT) single crystal has been reported with good thermal, optical and laser damage threshold properties. In order to find out a phase matching angle, a bulk crystal is required. The slow cooling cum seed rotation technique is the best one among various types of solution growth techniques. Generally, polar space group with noncentrosymmetric crystal system exhibits second order nonlinearity and piezo-electric property. Although some systematic studies have been carried out on IMLT crystal, device based characteristics such as phase matching angle have not been reported. Slow cooling cum seed rotation solution growth technique has been employed to grow IMLT crystal. Identification of crystal planes is also very important to perform dielectric tensor analysis and phase matching study. In the present investigation, we report bulk crystal growth, single crystal X-ray diffraction study, phase matching study and dielectric tensor analysis of imidazolium tartrate (IMLT) crystal.

2. Experimental

2.1 Crystal growth

The title material was prepared by dissolving imidazole and L-tartaric acid with an equimolar ratio in deionized water. The solution was stirred and subsequently kept for evaporation at 50 °C for several days. Then, the IMLT salt was collected and recrystallized several times in order to improve the quality of IMLT salt. Indigenously developed seed rotation setup with 3 rpm rotation speed was used for the growth of bulk crystal as shown in Fig.1(a). The saturated growth solution was prepared using recrystallized salts of IMLT and the same was placed in a constant temperature water bath. The temperature of the growth solution was maintained at 40 °C. A good quality seed crystal with well-developed facets was taken and placed on the bottom of the acrylic disc of the growth setup. The whole setup was immersed in the saturated growth solution. Subsequently, the temperature of growth solution was reduced by 0.1 °C per day as the growth progressed. It was found that the growth direction of IMLT crystal is found to be (010) plane. After a span of 30 days a good quality bulk crystal of IMLT was harvested as shown in Fig.1(b).

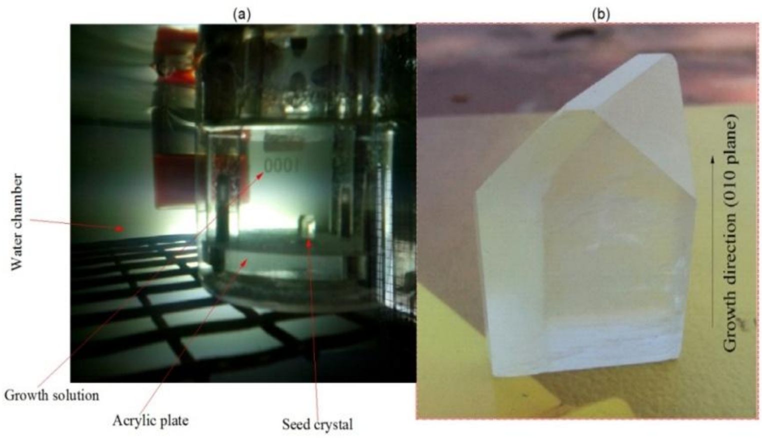

Fig. 1 (a) Growth setup and (b) as-grown IMLT crystal

3. Results and discussion

3.1 X-ray diffraction study

Bruker Kappa APEXII single crystal X-ray diffractometer using MoK$_\alpha$ (λ = 0.71073 Å) radiation was used to measure the cell parameters and morphology of IMLT crystal. The cell parameters of IMLT crystal are found to be a = 7.53 Å, b = 6.944 Å, c = 8.93 Å, β = 101.63°. Figure 2 shows the morphology of IMLT crystal. It has ten well developed facets with (-111), (11-1), (100), (-100), (001), (00-1), (-101), (10-1), (0-10) and (-131) planes. It is observed that 'b' axis has more growth rate than 'a' and 'c' axes.

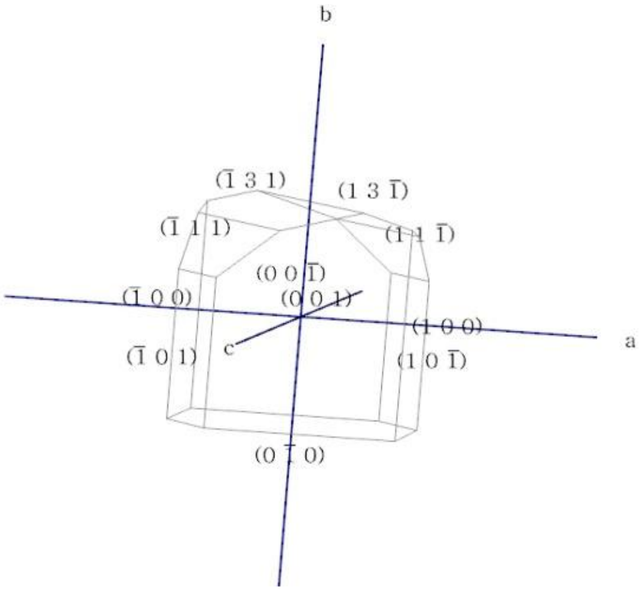

Fig.2 Morphology of IMLT crystal

3.2 Nonlinear optical study

An Nd:YAG laser with the wavelength of 1064 nm was used to find the phase matching angle and SHG output voltage of IMLT crystal. A laser beam of 1064 nm with 10 ns pulse width was exposed on the possible planes of IMLT crystal but the phase matching angle was found out along the direction of (0-10) plane with 42±0.2° (Fig.3). The laser beam with the size of 3.5 mm (diameter) was focused using a 20 cm focal length bi-convex lens and the focused beam size is 1.013 cm^{-2}. The laser damage threshold of IMLT crystal along (0-10) was found to be 11.7 mJ at 1 pulse per second and 9.44 mJ at 10 pulses per second. The relative SHG output of IMLT powder for particle size of 250 µm was found to be 4.3 times of KDP crystal [10-12].

3.3 Piezoelectricity

Piezoelectric measurement was carried out using a piezometer (High Precision Model PM300) at a tapping frequency of 110 Hz with tapping force 0.25 N). The piezoelectric measurement was carried out on a (001) plane of IMLT crystal. The direct (d_{33}) and transverse (d_{31}) charge coefficients of IMLT were measured to be 1 pC/N and 17 pC/N, respectively.

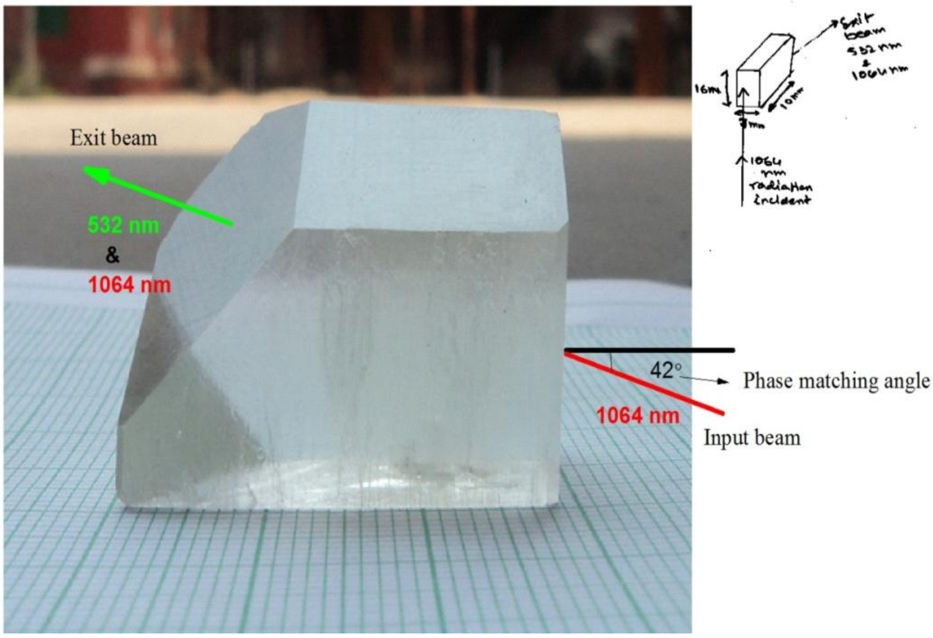

Fig.3 Occurrence of phase matching in IMLT crystal

4 Conclusions

In summary, a bulk crystal of IMLT was grown by the seed rotation cum slow cooling solution growth technique. Single crystal X-ray diffraction study revealed the morphology and the crystal system of IMLT crystal. The phase matching angle of IMLT is found to be 42±0.2° using a Nd:YAG laser. The relative SHG output of IMLT powder for the particle size of 250 µm was found to be 4.3 times of KDP. The direct (d_{33}) and transverse (d_{31}) charge coefficient

References

1. Chemla D S, Zyss J, "Nonlinear optical properties of organic molecules and crystals", Vol.1, Academic Press, London, (1987).
2. Subhashini R., Arjunan S., & Gunasekaran B. J. Phys. Chem. Solids, 2019; 135; 109077

3. Subahashini R., & Arjunan S. Opt. Laser Technol., 2018; 101; 248.
4. Sharma S. K, Verma S, Singh Y, Bartwal K. S, Cryst Eng Comm**,** 2013**;**15: 9955
5. Jagadesan A, Sivakumar N, Arjunan S and Parthipan G, Optical Materials 2020 ;109 ; 110285
6. Jagadesan A, Peramaiyan G, Mohan Kumar R, Arjunan, S. J. Cryst. Growth, 2015: 418; 153
7. C Chen, T Sasaki, R Li, Y Wu, Z Lin, Y Mori, Z Hu, J Wang, G Aka, M Yoshimura, Y Kaneda, Nonlinear Optical Borate Crystals: Principals and Applications, John Wiley & Sons, (2012).
8. Peramaiyan G, Pandi P, Bhagavannarayana G, Mohan Kumar R, Spectrochim Acta A Mol Biomol Spectrosc. 2012; 99:27.
9. Jagadesan, A., Sivakumar.N, Thinesh Kumar. R, Arjunan, S, Adv. Mater. Proc, 2021;3;122
10. Thomas S, Pati Y. A, Ramasesha S, Cryst. Growth Des. 2011; 11:1846.
11. Ok K.M, Chi E.O, Halasyamani, P.S, Chem. Soc. Rev. 2006;35:710
12. Kurtz S.K, Perry T.T, J. Appl. Phys. 1968;39:3798