

SYNTHESIS OF SOLUTION GROWN EFFICIENT SEMI ORGANIC NLO CRYSTALS FOR SHORT WAVE LENGTH GENERATION – L-ARGININE, L- HISTADINE WITH ADDITIVES.

G. Maruthi^{1,*} and R. Chandramani²

¹Department of Physics, East Point College of Engineering and Technology, Bangalore - 560049

²Department of Physics, Dayananda sagar college of Engineering, Bangalore, India.

*E-mail: gmaruthi72@gmail.com

ABSTRACT

Semi organic crystals containing organic material (L-Arginine/L-Histadine) with additives (KNO₃, CaNO₃ etc.) have been grown by aqueous solution method. Transparent crystals were obtained in a matter of 3 weeks. To know the role of additives on the SHG efficiency, KNO₃, CaNO₃ etc are added in appropriate ratio to the host material L-Arginine/ L-Histadine. Second harmonic generation (SHG) efficiency has been tested by Kurtz powder technique using Nd: YAG laser. KDP sample has been used as reference material. Role of additives on second harmonic generation efficiency has been investigated. In addition to SHG, other studies such as AC conductivity (σ_{ac}), dielectric constant (ϵ_r), dielectric loss ($\tan\delta$) have been carried out for the sample L-Histadine hydrochloride monohydrate (LHC).

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Key words: SHG, Dielectric constant, AC conductivity, dielectric loss.

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INTRODUCTION

Semi organic non linear optical (NLO) materials have attracted the interest of many theoretical and experimental researchers because of their appreciable nonlinearity, fast response and tailor made flexibility. Semi organic NLO crystals have been developed which retain the merits of organic material and overcome the shortcomings of organic materials¹⁻². Coherent blue and green light is important for certain application such as high resolution printing and signal processing³. These properties depend on dielectric and other properties⁴. Amino acids with inorganic salts are promising materials for second harmonic generation (SHG). These complexes are better alternative for KDP crystals in all respects. The literature survey reveals that growth and characterization has been done for pure LHC crystals. Present paper deals with the growth of L-Arginine and L-histadine with additives (Inorganic materials) by slow evaporation technique. The grown crystals were characterized by AC conductivity (σ_{ac}) dielectric constant (ϵ_r) and dielectric loss ($\tan\delta$) measurements.

EXPERIMENTAL

Crystal Growth

Single crystals were grown from supersaturated solution by slow evaporation technique. The solubility curves of L-Arginine and L-histadine in double distilled water were experimentally confirmed with the available one in the literature. The host material and additives were mixed in a molar ratio of 0.5:0.5. The supersaturated solution was stirred by using magnetic stirrer for homogenization. The containers of the solution were kept in a constant temperature bath for slow evaporation of the solvent. Crystals with good transparency were obtained in a time of 3 weeks.

SHG Measurement

The powder SHG efficiency measurement was made by using a set-up similar to the one devised by Kurtz⁵. A Q-switched Nd-YAG laser emitting 1064µm, 8ns laser pulses with spot radius of 1mm was

used in this experiment. The laser beam of input power 6.6mJ was passed through an IR reflector and then directed on the powdered sample. The emitted light from the sample was measured by the photo diode detector and oscilloscope arrangement.

Dielectric measurement

The grown crystals were grounded to proper thickness and were well polished. Each sample was electroded on either side with air-drying silver paste, so that it behaved like a parallel plate capacitor with crystal as dielectric medium. A 4275A, multi frequency LCR meter (Hewlett-Packard) was used to measure Capacitance, Dissipation factor, Impedance and phase angle. These data were used to calculate the real and imaginary parts of dielectric constant, real and imaginary parts of impedance and ac conductivity. The following formulae were used for calculating different parameters.

$$\epsilon_r = \frac{Cd}{A \epsilon_0}$$

$$\tan \delta = D \epsilon_r$$

$$Z' = |Z| \cos \theta$$

$$Z'' = |Z| \sin \theta$$

$$\epsilon_{r(\text{imag})} = \epsilon_r \tan \delta$$

$$\sigma_{ac} = \epsilon_0 \epsilon_r \omega \tan \delta$$

Where,

C- Capacitance of a capacitor; d- Plate separation; A- Area of plate; D- Dissipation factor; Z -Complex impedance; Z'-Real part of impedance; Z''-Imaginary part of impedance; θ -Phase angle; ω -Angular frequency; $\epsilon_{r(\text{imag})}$ – Imaginary part of dielectric constant; ϵ_r -Real part of dielectric constant; σ_{ac} -A.C. Conductivity.

RESULTS AND DISCUSSION

The SHG efficiency of different compounds are shown in table1. The SHG efficiency of pure LHC has reported to be three times that of KDP⁶. In the present investigation it has been found that it increased with the addition of KNO₃ but decreased with the addition of CaNO₃. For L-Arginine both the additives are acting as poison. Study of dielectric measurement of solids yields information about the electric field distribution within the solid. The variation of real and imaginary parts of dielectric constant at room temperature are shown in fig 1. and fig 2. respectively for LHC crystals containing KNO₃ and CaNO₃.

The dielectric constant is high at low frequencies and it decreases with increase in frequency. The electrical conductivity predominates in the low frequency range due to exchange of electrical charges at crystal surface. Whereas dielectric constant of a material is composed of four type's namely electronic, ionic, orientation and space charge polarisabilities. All these Polarisabilities are active in the low frequency region, later they freeze out one after the other. The variation of dielectric loss and AC conductivity are shown in fig. 3 and fig. 4 respectively. The low value of $\tan \delta$ in the high frequency region suggests that the grown samples are of moderately good quality⁷.

AC conductivity study is carried out to characterize the bulk resistance of the crystalline solids. Normally impedance measurements are made over a range of temperatures and frequencies and analyzed in the complex impedance plane.

Impedance is a more general term. In ac measurement the resistance R is replaced by the impedance Z which consists of two parts, written as $Z = Z' + jZ''$

Z is a phasor quantity-a rotating vector may be plotted in rectangular form / polar form / exponential form / trigonometric form. In rectangular form it takes the value as-

Real part of Z is, $Z' = |Z| \cos \theta$

Imaginary part of Z is, $Z'' = |Z| \sin \theta$

In polar form $Z(\omega) = |Z| \exp(j\omega)$

Where, $\exp(j\omega) = \cos \theta + j \sin \theta$

From the measurements real and imaginary impedances are evaluated as a function of frequency. The impedance plots (Nyquist plots) are shown in figure 5 and 6. The arcs can be interpreted by equivalent circuit model of the nature of the graph depicts the quality of the capacitor⁸.

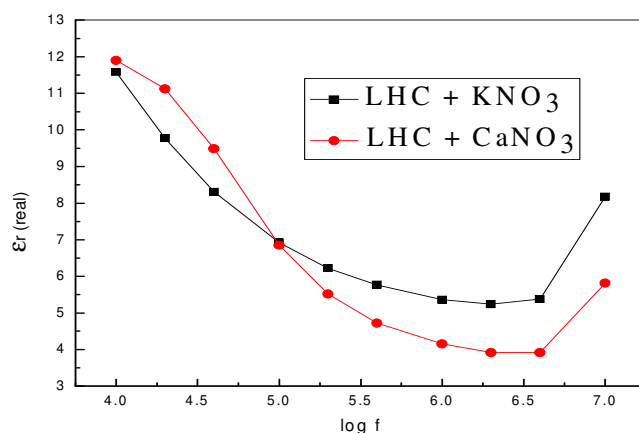


Fig.-1: Variation of real part of dielectric constant with frequency.

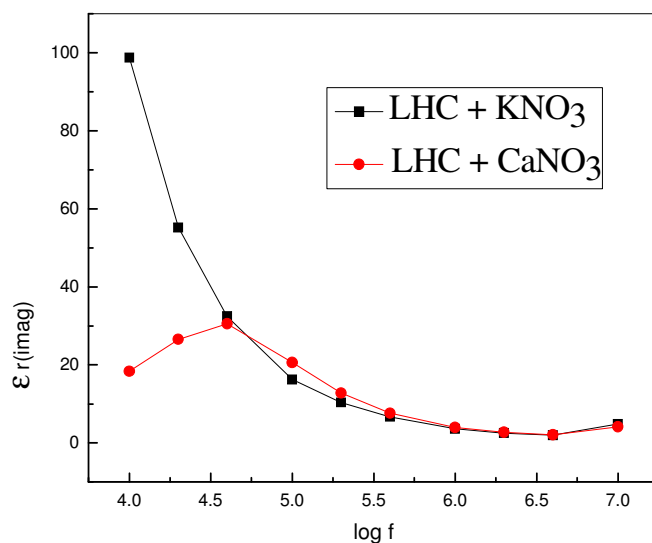


Fig.-2: Variation of imaginary part of dielectric constant with frequency.

Table-1: SHG efficiency of different compounds with respect to KDP.

Compound	composition	SHG Efficiency
L-Arginine	Pure	1.42
L-Arginine + KNO ₃	0.5+0.5	0.84
L-Arginine + CaNO ₃	0.5+0.5	1.32
LHC + KNO ₃	0.5+0.5	4.32
LHC+CaNO ₃	0.5 + 0.5	1.29

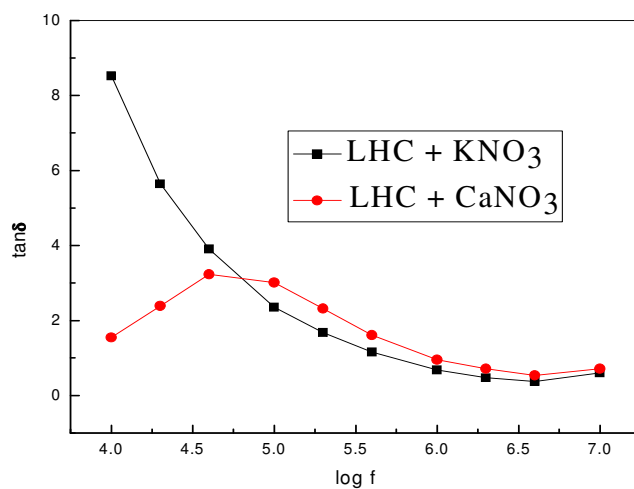


Fig.-3: Variation of dielectric loss with frequency.

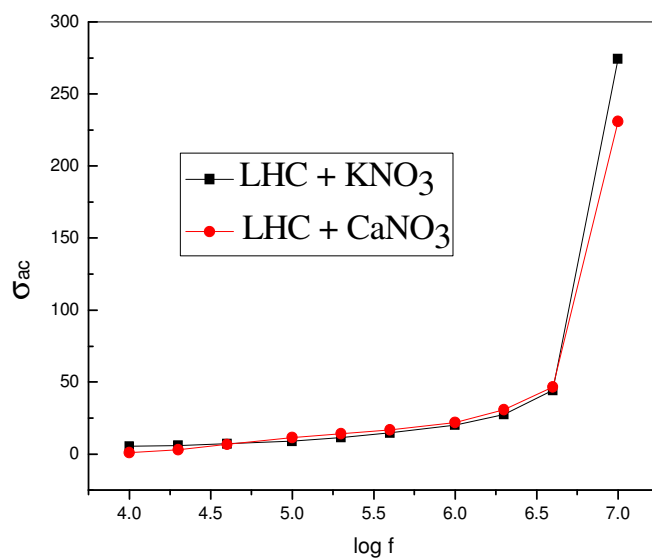


Fig.-4: Variation of ac conductivity with frequency.

CONCLUSIONS

Good quality semi organic crystals were obtained in 3 to 4 weeks. LHC with Potassium Nitrate has answered for very good SHG efficiency. Low value of $\tan \delta$ in grown crystals confirms good quality of the crystals. Nyquist impedance plots justify good quality as a capacitor.

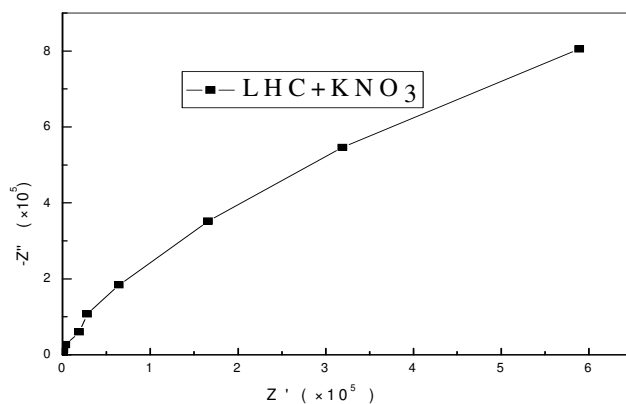


Fig.-5: Variation of real and imaginary parts of impedance for. LHC + KNO_3 crystal

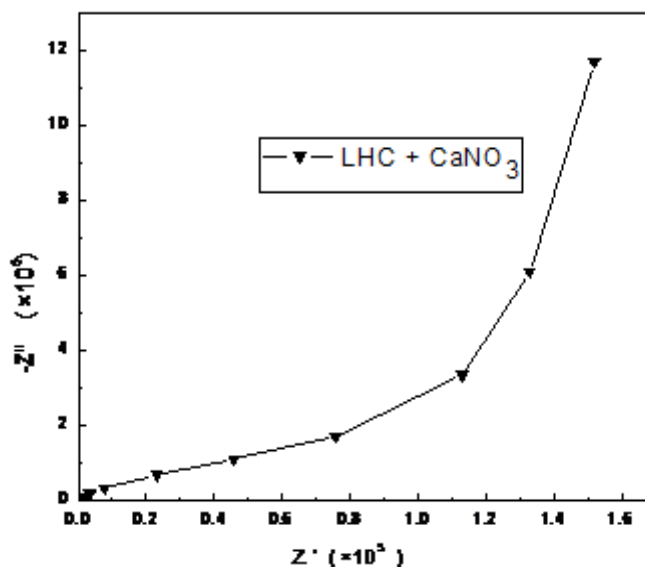


Fig.-6: Variation of real and imaginary parts of impedance for. LHC+ CaNO_3 crystal

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