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RESEARCH ARTICLE

DIELECTRIC STUDIES OF THIOUREA DOPED SULPHAMIC ACID SINGLE CRYSTAL

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ABSTRACT

Thiourea doped sulphamic acid single crystals (Th : SA) were grown by slow solvent evaporation technique at room temperature and their dielectric properties are studied. The variation of dielectric constants, dielectric loss and conductivity at different temperatures and frequencies in the applied field are measured and their electrical behaviors are analyzed. The activation energy of electrical process is also determined at various frequencies and temperatures.

INTRODUCTION

The electrical properties of the single crystals gain importance over the recent years, because of its application in the fabrication of new engineering devices. Dielectric studies is informative regarding the electrical response within the materials. The dielectric characteristics of a sample can be understood from the measurements of dielectric constant and dielectric loss. The dielectric constant of a material enumerates the nature of atoms , ions and its bonding in the material (Rajasekaran *et al*., 2002; Rajasekaran *et al*., 2001), whereas the dielectric loss is a measure of the energy absorbed by a dielectric (Rajesh *et al*., 2004; Brahadeeswaran *et al*., 1998). The present investigation is mainly on the study of dielectric properties of (Th : SA) single crystals.

Synthesis and growth

(Th : SA) salt was synthesized by dissolving (AR grade) thiourea and sulphamic acid in the molar ratio 1:1 in double distilled water. (Th : SA) was synthesized and the product was purified by repeated recrystallization, typically thrice from double distilled water. The 100 ml saturated solution was kept in a beaker covered with perforated polyethylene lid at room temperature. The crystals were grown by low temperature solution growth, using solvent evaporation method.

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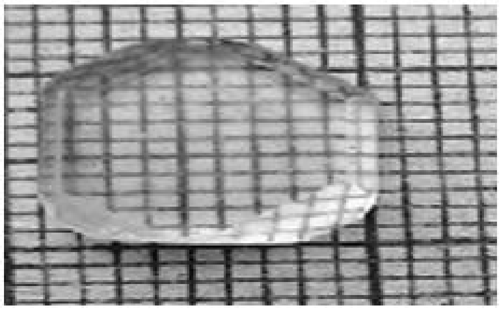

Fig.1. (Th : SA) single crystal

In a period of 3 to 4 days, seeds of (Th : SA) single crystals were formed due to spontaneous nucleation as shown in Fig.1 The crystals with good transparency, well shaped and free from defects were selected and its electrical properties were studied by the following experiments.

Characterization

The different characterization studies such as, single crystal xrd, powder xrd, morphological, optical, thermal and NLO studies have been already reported *(*Kannan *et al., 2013)*. However no systematic efforts were made to study the electrical properties of the title crystal. Hence the present work is aimed to reveal the electric properties, such as variation of dielectric constant, dielectric loss, ac conductivity and activation energy with temperature and the frequency of the input signal for (Th : SA) single crystal.

Experiment

The dielectric constant and loss of grown (Th : SA) single crystal were calculated using HIOKI 3532 LCR HITESTER in the frequency region 50 Hz to 50 MHz. The samples were pelletized and a pellet of uniform dimension was placed between the two copper electrodes, which acts as a parallel plate capacitor. Silver paint was coated on the surface of the sample in order to make firm electrical contact. The capacitance, impedance and loss were measured for the applied frequency varies from 50 Hz to 50 MHz for three different temperatures 303 K, 353 K, and 393 K respectively for thiourea sulphamate single crystal (Dhaumane *et al*., 2006).

Dielectric constant

The dielectric constant was calculated using the relation $\varepsilon_r = C d / \varepsilon_o A$ where C is the capacitance in F, d the thickness in m, A the area of the crystal in m^2 and ε_o is the absolute permittivity in free space having a value of 8.854×10^{-12} farad/meter. The dielectric constant of a substance is a property of the constituent ions (Gaffar *et al*., 2006). In general, if electrode effects are neglected the four major components contributing to the dielectric constant are the extrinsic nature of the material, the electronic polarisability, the ionic polarisability and the deformation of ions.

Dielectric loss

The dielectric loss is calculated using the formula $\tan \delta = 1/\omega RC$. The factor $\tan \delta$ is a measure of the energy absorbed by dielectrics. It is known that is a capacitor the dielectrics usually has a resistance R (assuming R to be very large) and impedance $Z = 1/\omega C$ which are related to the phase angle.

AC conductivity

The ac conductivity $\ln \sigma_{ac}$ is calculated by substituting the values of the dielectric constant ε_r and the dielectric loss $\tan \delta$ (measured directly from the instrument) in the formula $= 2\pi f \varepsilon_o \varepsilon_r \tan \delta$ where ε_o is the absolute permittivity of free space having the value of 8.854×10^{-12} farad/metre and f is the frequency in hertz (Narasimha *et al*., 1988).

RESULTS AND DISCUSSION

Fig. 2 shows the variation of dielectric constant with the frequency.

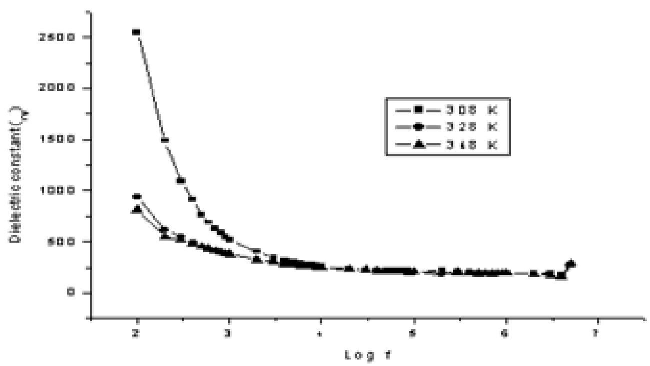

Fig. 2. Variation of dielectric constant as a function of frequency at various temperatures

The surface charge polarization, which depends on the frequencies hence in the graph large value of dielectric constant at lower frequency may be due to interfacial or space charge polarization (Narasimha *et al*., 1988; Rao and Smakula, 1965) arising at the grain boundary interfaces. The polarization and hence the dielectric constant in a medium can be considered as a measure of the energy associated with the electrostatic binding (Balrew *et al*., 1984). The higher the value of the dielectric constant at low frequency, lower the electrostatic binding. The dielectric constant is temperature dependent due to the change in the position configuration of atoms or molecules with the rise of temperature. This leads to the change in the entropy or free energy of the system which can be related to the permittivity (Goswami, 1996). From the Fig 3, it is seen that the dielectric constant decreases with increase in temperature and almost remains constant at higher temperatures.

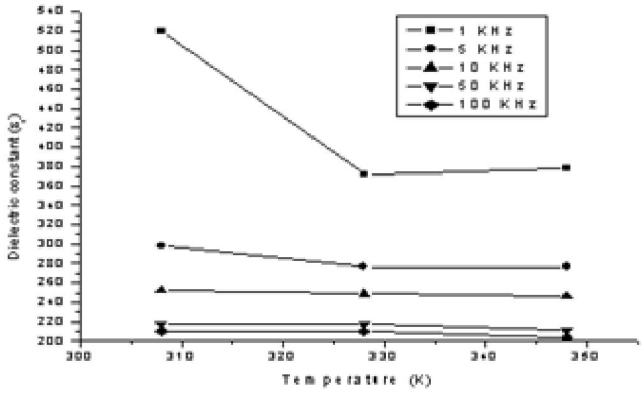

Fig. 3. Variation of dielectric constant as a function of temperature at various frequencies in Th : SA single crystals

A plot is drawn between log frequency and dielectric loss for various temperatures and a plot is drawn between temperature and dielectric loss for various frequencies and the resulting plots are shown in Fig 4 and Fig 5. From the Fig 4, it is understood that the dielectric loss decreases with increase in frequency. The low dielectric loss confirms the purity of the synthesized sample. The characteristic low dielectric value for the grown crystals suggests that the crystals possess enhanced optical quality with lesser defects and this parameter is of vital importance for various nonlinear optical materials and their applications (Ramesh *et al*., 2007).

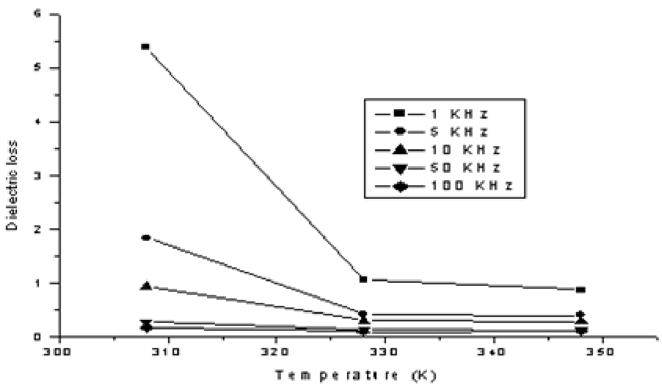

Fig. 4. Variation of dielectric loss as a function of frequency in Th: SA single crystals

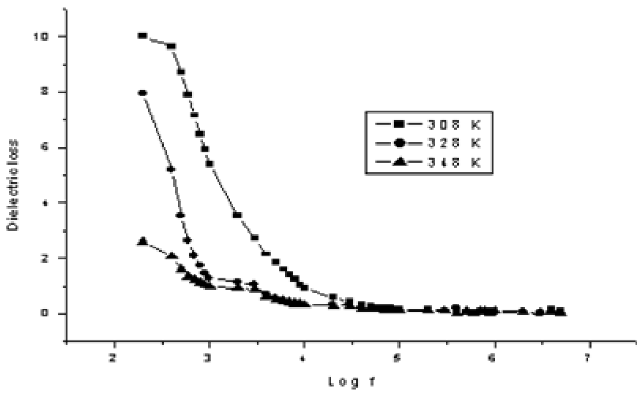

Fig. 5. Variation of dielectric loss as a function of temperature at various frequencies in Th : SA single crystals

From the Fig 5, it is seen that the dielectric loss also decreases with increase in temperature and almost remains constant at higher temperatures A plot is drawn between log frequency and ac conductivity for various temperatures and a plot is drawn between temperature and ac conductivity for various frequencies and the resulting plots are shown in Fig 6 and Fig 7. From the Fig 6 it is concluded that the ac conductivity increases with increase in frequency.

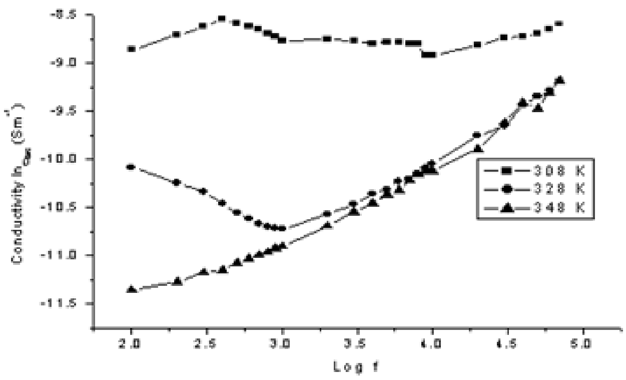

Fig. 6. Variation of ac conductivity as a function of frequency at various temperatures in Th : SA single crystals

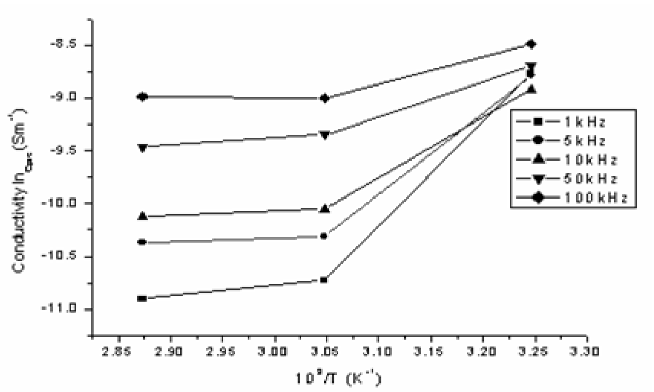

Fig. 7. Variation of ac conductivity as a function of temperature at various frequencies in Th : SA single crystals

From fig 8, it is seen that the ac conductivity decreases with increase in temperature and almost remains constant at higher temperatures. A plot is drawn between conductivity versus 1/T is shown shown in Fig 7. The activation energies at various frequencies of 50 kHz, 100 kHz, 1 MHz, 5 MHz and 10 MHz are calculated from the slope of the graph drawn between conductivity versus 1/T as 0.1039, 0.1333, 0.1764 and 0.4258 eV.

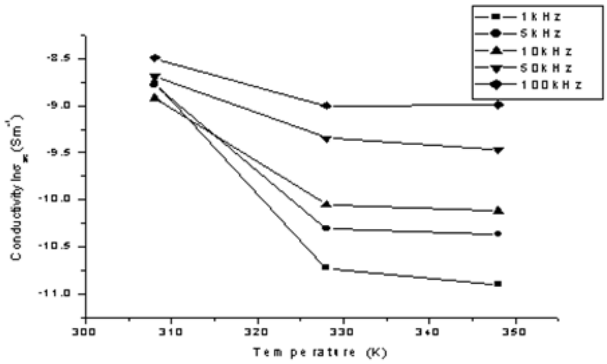

Fig. 8. Variation of ac conductivity as a function of 1/T in Th : SA single crystals

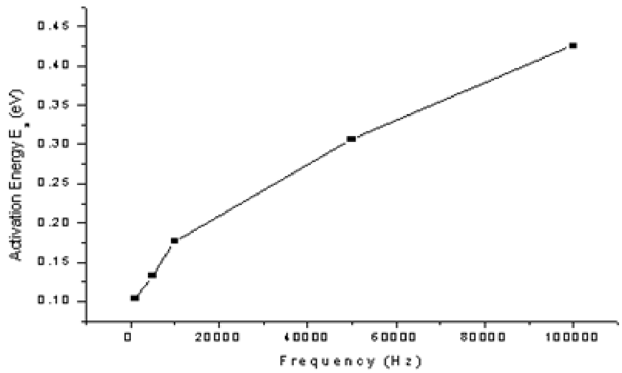

Fig. 9. Variation of activation energy as a function of frequency in Th : SA single crystals

The slope changes at different regions of temperature indicates a variation of activation process, that is, activation energy due to thermal excitation and consequent jumping of electrons to the conduction band. The plot drawn between frequency and activation energy is shown in Fig 9 and the trend shows that the activation energy increases with increase in frequency.

Conclusion

The variation of dielectric constant, dielectric loss, ac electrical conductivity and activation energy as a function of temperature and frequency of (Th : SA) single crystal was measured for the first time. These variations are observed along [100] direction. The results showed that the dielectric constant and dielectric loss of the grown crystal decreases with increase in frequency and temperature respectively whereas the dc electrical conductivity measurements showed that the conductivity increases with the frequency and decreases with the increase in temperature.

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