INTERNATIONAL JOURNAL OF INNOVATIVE RESEARCH & DEVELOPMENT

ISSN: 2278 – 0211 (Online)

Dielectric Studies On Pure And Metal Doped KDP Single Crystals

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Abstract:

Single crystals of pure and mercuric (II) chloride doped KDP have been grown by slow evaporation method. Dielectric constant and dielectric loss have been obtained as a function of frequency between 1 KHz and 5 MHz and temperature range between 353 K and 403 K. The dependence of tan δ, ε and σ_{ac} on temperature and frequency of the applied field (50 Hz–5 MHz) is established. Both dielectric constant and dielectric loss decreases with increase in frequency. Impedance plot has also been drawn at four different temperature and results are discussed in the paper.

Keywords: *KDP, dielectric loss, dielectric constant, Impedance etc*

1.Introduction

The study of dielectric properties is concerned with the storage and dissipation of electric and magnetic energy in materials. It is important to explain various phenomena in electronics, optics and solid state physics. The term dielectric applies to the material properties governing the interaction between matter and electromagnetic field. Induced or permanent electric polarization or magnetization of matter as a function of static or alternating electric, magnetic or electromagnetic field constitutes the dielectric properties of the material. The dielectric constant is one of the basic electrical properties of solids. Various polarization mechanisms in solids such as atomic polarization of the lattice, orientational polarization of diploes and space charge polarizations can be understood very easily by studying the dielectric properties as a function of frequency and temperature for crystalline solids [1-3]. These investigations help in detecting the structural phase transitions taking place in solids when abrupt changes in dielectric properties are observed. Particularly, the presence of a dielectric between the plates of a condenser enhances the capacitance. The effect makes material with dielectric constant useful in capacitor technology. Microelectronics industry needs low dielectric constant (ε_r) materials as an interlayer dielectric [4].

The magnitude of the dielectric constant depends on the degree of polarization charge displacement in the crystals. The electronic exchange of the number of ions in the crystals gives local displacement of electrons in the direction of applied field that gives polarization. Ferroelectric domains are areas of such local dipole alignment with an associated net dipole moment and net polarization. In presence of an applied electric field, domains that are aligned with the direction of the field will grow at the expense of the less well aligned domains. Thus the observed enhancement in the dielectric constant at low frequency could be attributed to the multidomain state of the proline doped sample. As the frequency increases, a point will be reached where the space charge cannot sustained and comply with the external field giving rise to diminishing values of dielectric constant. Above this frequency the domain wall motion contribution to dielectric constant was nearly the same for pure and doped crystals [5].

2.Materials And Methods

AR grade samples of Pottasium dihydrogen orthophosphate (KDP) and Mercuric (II) chloride were used. 200 ml saturated solution of KDP in distilled water is prepared at first and 100 ml saturated solution is poured in two different beakers and named as 1 and

2. To that saturated solution, 0.2% of mercuric (II) chloride is added to beaker 2 and stirred well for nearly 5 hours. The stirred solution is filtered and allowed to evaporate slowly. Within a week, transparent crystals were produced.

3.Dielectric Measurement

Suitably cut and polished section (001) of pure and mercuric (II) chloride doped KDP crystal were subjected to dielectric studies using a HIOKI model 3532-50 LCR HITESTER with a conventional two terminal sample holder. The sample was electroded on either side with air-dying silver paste so that it behaves like parallel capacitor. The studies were carried from 353 K -403 K for frequency varying from 50 Hz to 5 MHz.

4.Result And Discussion

Figure 1 and 2 shows the variations of dielectric constant with log frequency for pure KDP and mercuric (II) chloride doped KDP. The dielectric constant is calculated by using the formula

$\varepsilon' = C\ t / \varepsilon_o\ A$ ---------- (1)

Where C is capacitance (F), t is the thickness (m), A the area (m^2), ε_o is the absolute permittivity in the free space having a value of $8.854 * 10^{-12}$ $Fm^{-1.}$

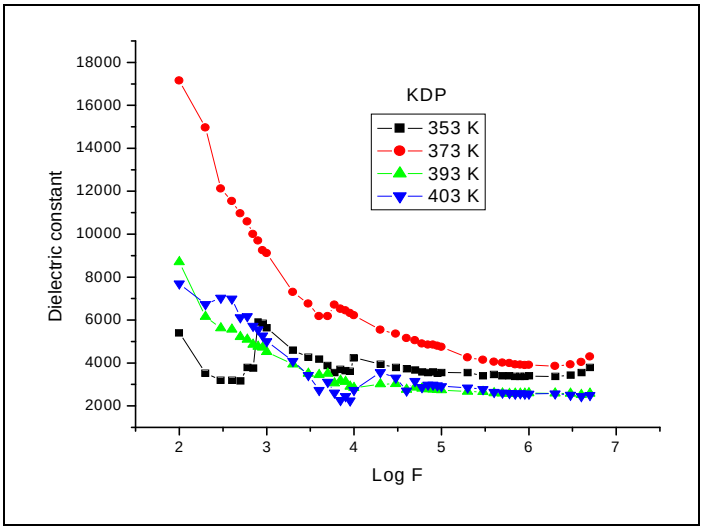

Figure 1: Variation of dielectric constant with log frequency for pure KDP

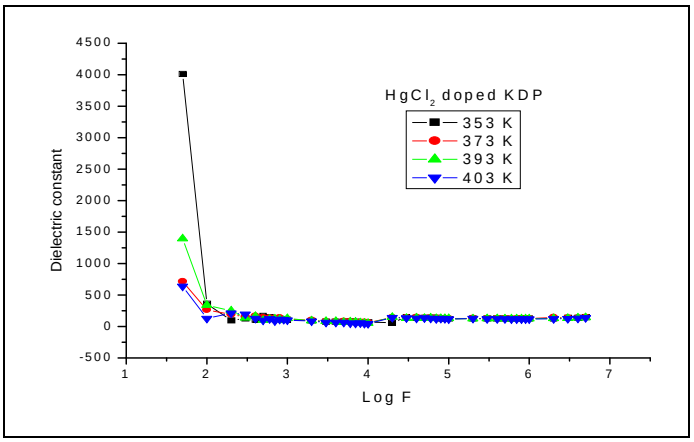

Figure 2: Variation of dielectric constant with log frequency for Hgcl$_2$ doped KDP

It is seen from the figure 1 and 2, that the dielectric constant decreases with increasing frequency at almost all temperatures and appears to attain saturation at high frequency range of 50 KHz and above. The decrease in the dielectric constant of the material at low frequencies may be attributed to the contribution of the electronic, ionic, orientation and space charge polarization which depend on the frequencies [1,6.At low frequencies all the four polarizations are active. It is also seen that dielectric constant increases with increasing temperature. The high value of dielectric constant is attributed to high ionic conductivity [7]. Most of the solid electrolytes have higher dielectric constant [8]. Higher values of dielectric constant at higher temperature may be due to the thermal excitation of atoms about their lattice points and these would have happened due to disorder at the lattice. Space charge contribution to polarization may be attributed to the purity of the crystal. Dielectric constant at low frequencies can be compared to optical frequencies[9]. This leads to minimization of the phase mismatch between optical and electrical pulses in high speed traveling wave devices. However, the low values of dielectric loss suggest that the grown crystals possess good optical quality which is the essential parameter for the non linear optical material for their applications. [10]. Hopping of charge carriers would have happened in the lattice sites and this has happened because of thermal excitations of atoms about their lattice points [11].

At low frequencies the mobile charges, usually impurity ions diffuse under the influence of the applied electric field up to the interface and build up the surface charge until the applied field reverses with the alternating frequency of the alternating field and this cannot follow the field variations at very high frequencies [12].

The variation of dielectric loss as a function of frequency at different temperatures for pure and $Hgcl_2$ doped KDP is depicted in figure 3 and 4. And it is found that the same trend is followed as that of dielectric constant. It is also observed that the higher dielectric loss occurs at higher temperatures and at lower frequencies. This is understandable from the fact that at lower frequencies the trend is due to space charge polarization and at higher temperatures the trend may be due to macroscopic distortion in the charges [12].

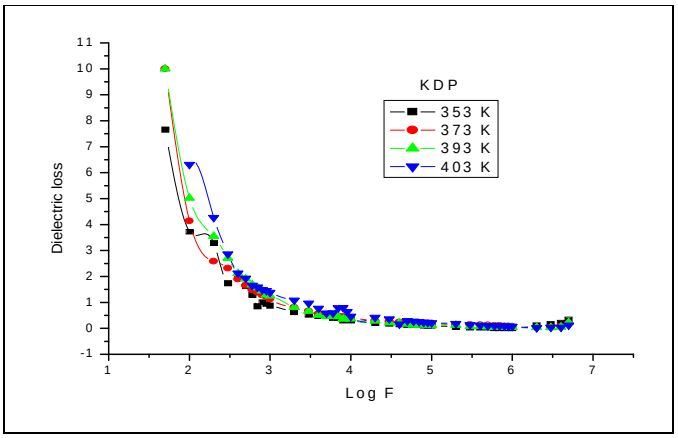

Figure 3: Variation of dielectric loss Vs log f for pure KDP

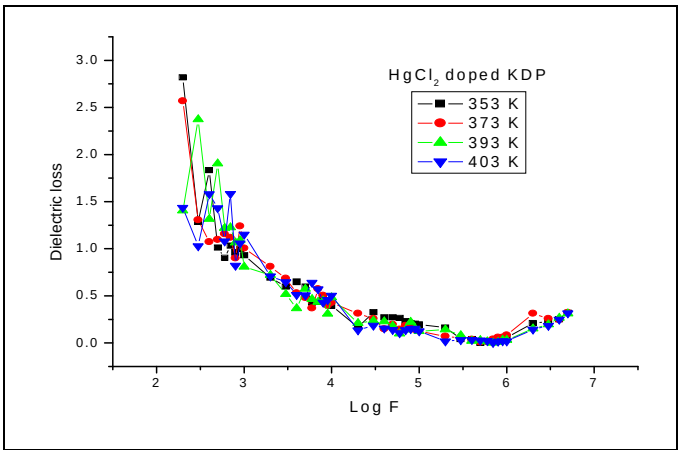

Figure 4: Variation of dielectric loss Vs log f for $HgCl_2$ doped KDP

It is found to have a relatively low tan δ value, indicating that it possesses lesser number of electrically active defects, which is a vital parameter in electro-optic device fabrications. In normal dielectric behavior, the dielectric constant remains almost a constant at high frequency, because beyond a certain frequency of electric field, intrawell

hopping becomes prominent, and the charge carriers do not get enough time for long range hopping before field reversal [1].

It is also seen that tan δ remains almost constant at certain frequency ranges over a certain range of temperature and this happens in our case at 10 KHz, 100 KHz and 1 MHz. In the case of 1 KHz and 5 MHz there is sine wave like variation as temperature is increased. Barring the value at 5 MHz the general trend can be assumed as the value of tan δ is found to increase with increasing temperature and decreasing frequency. The variation of dielectric loss with respect to temperature and frequency may be due to space charge polarization [13]. Further the space charge polarization can be explained through Shockley-Read mechanism [9]. For low and middle order frequencies and at high temperature the impurity ions in the bulk crystal matrices capture the surface electron, causing the space charge polarization at the surface. The electron capture process increases with increase in temperature. This type of surface polarization can be accounted by statistical method and that is the reason for explaining the above results through Shockley- Read mechanism [9]. By this mechanism it is given that at low and middle order frequencies and at higher temperatures the impurity ions in the bulk crystal matrices capture the surface electron, causing the space charge polarization at the surface. The electron capture process increases with increase in temperature. By this mechanism one can presume that the loss tangent increases with increasing temperature and at low frequencies [14].

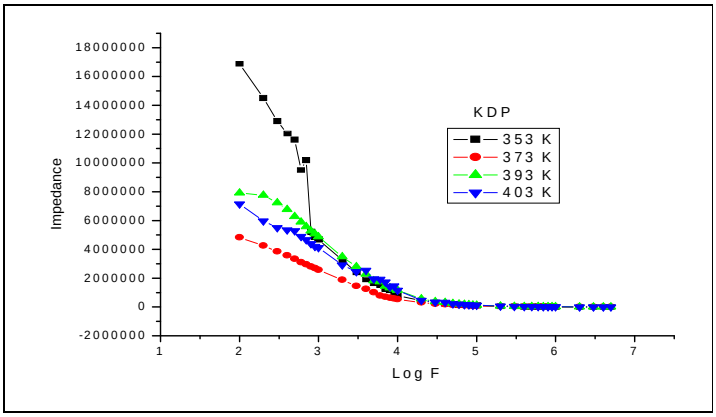

Figure 5: Impedance plot of pure KDP

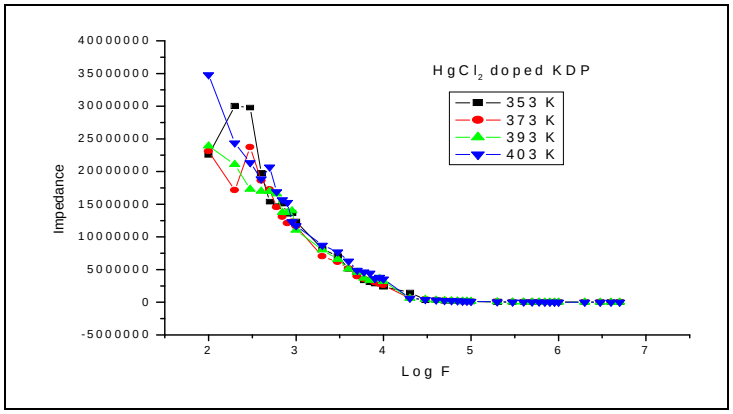

Figure 6: Impedance plot of $HgCl_2$ doped KDP

Figure 5 and 6 shows the impedance plot for pure and $HgCl_2$ doped KDP. It is clear that impedance is high at low frequencies for both pure and metal doped KDP and decreases with increase in frequency. Also a significant variation in impedance occurs for different temperatures.

It is seen that a.c. conductivity is governed by the presence of a small number of free charges which result in small leakage or conduction currents and by the displacement of bound charges that give rise to polarization or displacement currents in the solid state dielectrics whereas in the d.c. conductivity there is no contribution from localized charges. Further ionic conduction plays a major role at higher temperatures for both a.c and d.c conductivity because at higher temperatures some ions detach from the sites of crystal lattices[15] However, at lower temperatures mostly weekly bound ions, particularly impurity ions, are free to drift as already discussed. At low frequency space charge polarization is dominant mechanism in the transport processes which is absent at high frequencies. The space charge polarization decreases with increase in frequency due to inertia of the ions to follow the variation in field.

The variation in resistivity and conductivity with the frequency for the title crystal is shown in Figure 7-10. The a.c resistivity and a.c conductivity were calculated using the relation

$\rho=A/2\pi fCd$ and $\sigma_\rho=1/\rho$ (2)

Where C is the capacitance, d is the thickness, A is the area of the crystal and f is the frequency of the applied field.

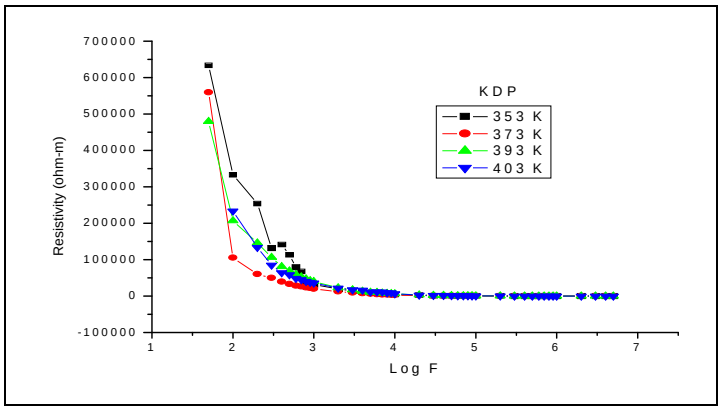

Figure 7: Variation in resistivity Vs log frequency for pure KDP

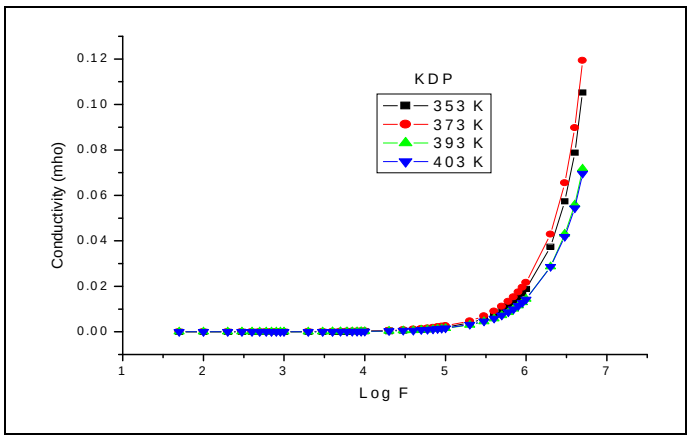

Figure 8: Variation in conductivity Vs log frequency for $HgCl_2$ doped KDP

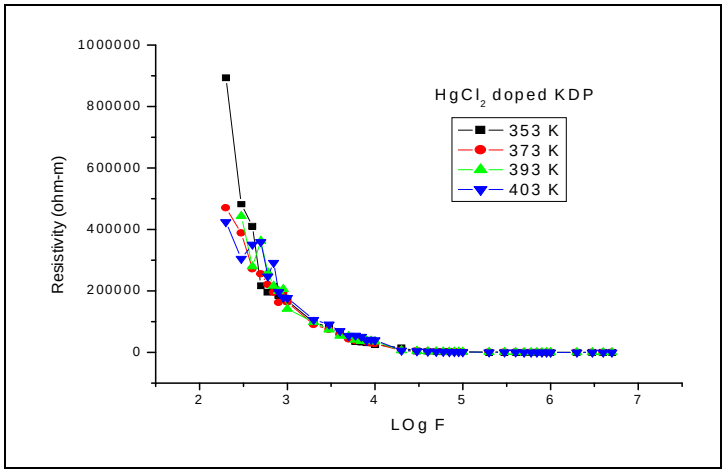

Figure 9: Variation in resistivity Vs log frequency for pure KDP

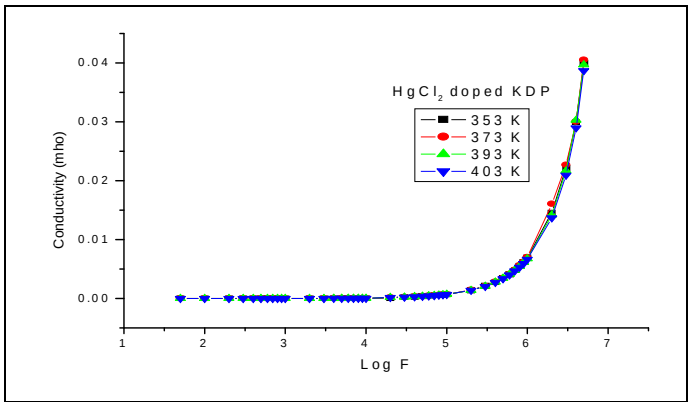

Figure 10: Variation in conductivity Vs log frequency for $HgCl_2$ doped KDP

It is observed that a.c resistivity decreased rapidly as frequency increased. Obviously reverse trend was observed for a.c conductivity of the grown crystals which is considered to be a normal dielectric behavior. Hence, conductivity increases with increases in temperature is due to the temperature dependence of the proton transport [16].

5.Conclusion

The dielectric constants (ε' and ε'') , dielectric loss (tan δ) and conductivity (σ_{ac}) of grown crystals are strongly dependent on temperature and frequency of the applied ac field, the variation depends on the ranges of temperature and frequency. The dielectric constant and dielectric loss decreases with increasing frequency and higher values of dielectric constant occurs at higher temperature. The rate of variation of imaginary dielectric constant (ε") with temperature is strongly dependent on temperature and frequency of the applied field. The conductivity increases with temperature. From all those analysis, it can be concluded that both pure and metal doped KDP is not only a potential Non linear Optical material but also a promising low ε_r value dielectric material, expected to be useful in the microelectronics industry. The encouraging dielectric properties of the crystal indicate the suitability of this crystal for photonics device fabrication. It is concluded that the value of dielectric constant decreases sharply in the low frequency range and attains the very low value at all frequencies for all temperatures.

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