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Effect of Nickel Oxide Nanoparticles on Dielectric and Optical Properties of Nematic Liquid Crystal

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Abstract. In the present paper, we have studied the improvement in dielectric and optical properties of nematic liquid crystal (NLC) by doping of nickel oxide (NiO) nanoparticles. We have observed the dielectric and optical properties of pure and doped cells in order to understand the influence of NiO nanoparticles in the pure NLC. The experimental results have been analyzed through dielectric spectroscopic and optical texural methods.Detailed studies of dielectric parameters such as dielectric permittivity, dielectric loss and dielectric loss factor as a function of frequency with temperature were carried out. It has been observed that on doping the nanoparticles in NLC, the value of dielectric parameters (dielectric permittivity, dielectric loss and dielectric loss factor) decreases. The impedance and resistance of both pure and nanoparticles doped NLC cells were studied and found that for doped NLC, these parameter have low value. In addition to this, optical textures of the pure and doped samples have also been observed with a polarizing optical microscope at room temperature. All the results *i.e.* related to the investigation of dielectric and electro-optic properties have been explained by using existing theory of NLC.

INTRODUCTION

Delicate, beautiful, mysterious, dynamic are words that describe the rich class of liquid crystals (LCs). In recent years, composite based on LCs and nanoparticles have been extensively investigated because of their unique dielectric and electro-optic properties for technology, industry, medical, novel display and non-display applications.[1]The nematic phase is the least ordered phase of the LCs. In nematic phase the molecules have no positional order but tend to orient along a preferred direction, known as director ($\hat{n}$).[2] Nematic liquid crystals (NLCs) have received much attention and deep interest in the recent years because of electro-optic applications, optical switches, LC displays (LCDs) as well as non-display applications. The key factor for all possible application is the alignment of LC molecule (*i.e.* director) on the substrate.[3,4]

The insertion of nanoparticles influences the properties of LCs and attracts scientific interest. Nanoparticles are considered to be the building blocks for nanotechnology. The dielectric and electro-optic properties of LCs can be enhanced on the dispersed materials, such as nanoparticles, polymers, dyes, carbon nanotubes etc.[5,6] 4-cyano-4'- pentylbiphenyl (5CB) exhibit nematic mesophase and is one of the best known liquid crystalline materials. 5CB, as well as other members of the nCB homogeneous series, is important from the point of view of applications due to the possession of a strong dipole moment, good chemical stability and convenient range of the nematic phase.[7] The selection of nanomaterials as dopants in LCs is important to enhance the properties of the LC material. Shape, size and concentration of nanomaterials have significant effect on the properties of the composite system.Many researchers are trying to improve the properties of LCDs by doping them with non-mesogenic materials. The properties of the pure NLCs can be optimized by the addition of nanoparticles. The addition of the nanoparticles may produce defects in nematic phase and breaks its continuous rotational symmetry. The properties, which can varied by dispersion of nanoparticles in the pure NLC, are dielectric permittivity, dielectric loss, dielectric loss factor ($\tan\delta$), impedance, resistance.[8]

In the present study, we have investigated the effect of NiO nanoparticles on dielectric and optical properties of NLC. We have observed the change in dielectric properties along with the change in optic

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parameter. The present paper is an effort to optimize dielectric and optic properties of pure NLC by doping NiO nanoparticles. The dielectric and optical properties have been observed for pure and nanoparticle doped NLC. The frequency and temperature dependence of the above mentioned parameters has also been discussed with the help of existing theories of NLC. It was observed that the doping of 1% of nanoparticles has causedvariation in almost all the properties of the NLC under consideration.[8] Thus we have prepared a LC mixture of 5CB and NiO nanoparticles to improve its dielectric and optic properties.

EXPERIMENTAL DETAIL

Liquid Crystal and Nanoparticles Used

The material studied in present work is commercially available NLC material namely 5CB. The chemical formula of 5CB is$C_{18}H_{19}N$ and itis the most common NLC. Fig.1 represents the molecular structure of 5CB. The NiO nanoparticles were used in the present study. The weight concentration of NiO nanoparticles was 1%.

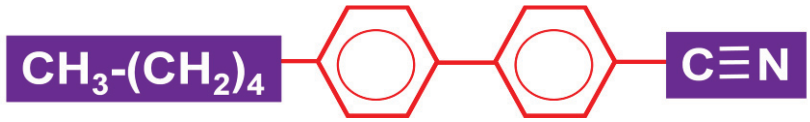


FIGURE 1. Molecular structure of nematic liquid crystal (5CB) used.

Preparation for Sample Cell

The dielectric and optical study of nanoparticle doped NLC were conducted on homogeneously aligned sample cells. The LC sample cells have been fabricated by using highly conductive and optically transparent sputtered indium tin oxide (ITO) glass substrates. The desired square electrode patterns were obtained by using photolithographic technique. The homogeneous alignment of LC sample cells has been achieved by using a conventional rubbed polyimide technique. The substrates were then placed one over another to form a capacitor.The cell thickness was maintained by placing a Mylar spacers between the two substrates and then sealed with UV sealant. The assembled cells were filled with pristine LC and nanoparticles doped LC doped by capillary action. After filling the cell, the alignment of the samples was checked under the crossed position of a polarizing microscope.

Apparatus and Measurement

The parameters such as dielectric permittivity, dielectric loss, dielectric loss factor, impedance and resistance of pure and doped cells have been measured using impedance analyzer (Wayne Kerr, 6540 A, U.K) and Temperature Controller (Julabo F-25 HE) was used for controlling the temperature. The sample holder containing the sample cell was kept thermally isolated from the external sources. The optical micrographs of the pure and doped sample cells were mounted on a polarizing microscope (Carl Zeiss Axioskop Ax-40).

RESULTS AND DISCUSSION

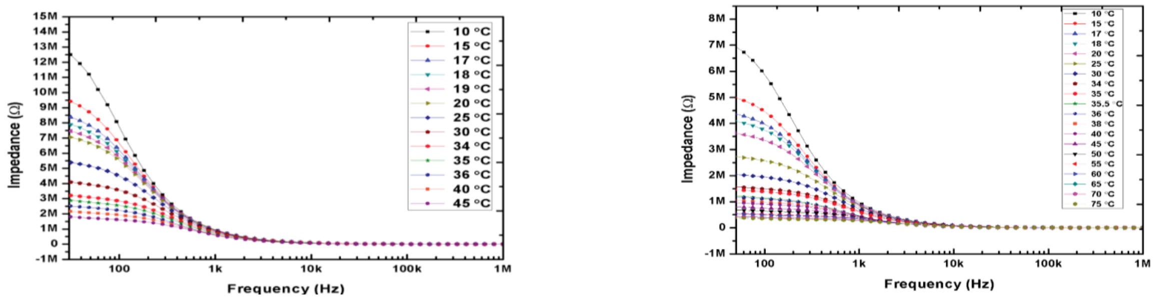
Impedance Measurements

To describe the electrical properties of materials and their interfaces with electronically conducting electrodes, an impedance spectroscopy method is generally used. Electrical measurements to evaluate the electrochemical behavior of electrode and electrolyte material are usually made with cell having two identical electrodes applied to face of a sample.[9] Fig. 2 shows the impedance (Z) of the samples as function of frequency at different temperatures.The complex impedance defined as

$$Z = R + iX \tag{1}$$

where R is the real part, *i.e.* resistance and X is the imaginary part, *i.e.* reactance.

For the pure NLC it has been observed that the Z decreases with the increase in the frequency. The Z has been found higher values at the low frequency, and attains a constant value at higher frequency regime. In low frequency region the Z has strong frequency dependence. This fact arises due to high resistivity in low frequency region. The high resistivity is due to effectiveness of the resistive grain boundaries and displays frequency independent behavior in high region. For the doped NLC it has been observed that the complex Z decreases with the increase in the frequency. Since the Z has strong frequency dependence in low frequency region. The variation of complex impedance with frequency shows almost similar behavior for pure and nickel oxide doped nanoparticles NLC but the values of Z are found to be lower for doped NLC. The variation of Z with temperature has been also analysed for pure and doped NLC. The Z decreases with increase in the temperature for pure and doped NLC. The Z is dependent on the temperature. This is due to the anisotropy of



(a) (b)

FIGURE 2. Behavior of impedance as a function of log of frequency for (a) pure nematic liquid crystal (5CB) and (b) NiO nanoparticles doped nematic liquid crystal (5CB) at different temperatures the samples.[10,11,12]

Dielectric Measurements

The study of dielectric permittivity and dielectric loss with frequency at different temperature provides the information regarding to polarization mechanism in solid, nature of an atom, ion and their bonding in the materials. The complex dielectric constant of the LC is defined as[13]

$\varepsilon^* = \varepsilon' - i\varepsilon''$, (2)

where $\boldsymbol{\varepsilon'}$ is the real and $\boldsymbol{\varepsilon''}$ is the imaginary part of the dielectric permittivity. The real part is described as

$$\boldsymbol{\varepsilon'} = \boldsymbol{C}\frac{\boldsymbol{d}}{\boldsymbol{\varepsilon_0 A}}, \qquad (3)$$

where d is the thickness, A is the effective area, $\boldsymbol{\varepsilon_0}$ is the permittivity of free space. The imaginary part is expressed as

$$\boldsymbol{\varepsilon''} = \boldsymbol{\varepsilon'} \tan \boldsymbol{\delta}, \qquad (4)$$

where $\tan\delta$ is dielectric loss factor. The spectra of real and imaginary parts are called dispersion and absorption curves.[12]

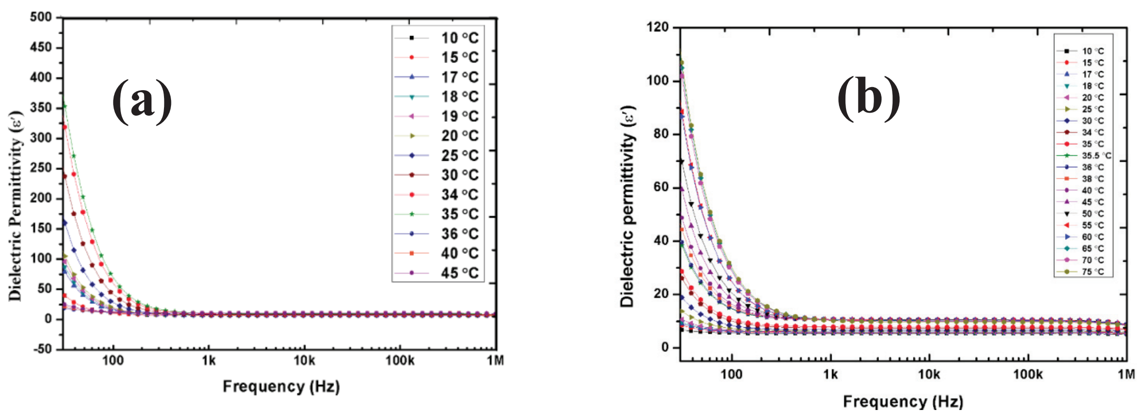


FIGURE 3. Behavior of dielectric permittivity (ε')as a function of log of frequency for (a) pure nematic liquid crystal (5CB) and (b) NiO nanoparticles doped nematic liquid crystal (5CB) at different temperatures**.**

In the present study, two types of cells were made, one for pure NLC material and second for NiO nanoparticles doped NLC system. Fig**.**3 shows the variation of dielectric permittivity ($\boldsymbol{\varepsilon'}$) with frequency at different temperatures for pure and nickel oxide nanoparticles doped NLC. At low frequency the$\boldsymbol{\varepsilon'}$is found to be decrease quickly, and then slowly as frequency increases, and finally the $\boldsymbol{\varepsilon'}$becomes constant with further increase in frequency. At 35 ℃, the value of $\boldsymbol{\varepsilon'}$ for pure NLC is 351.53 at 31.81 Hz and decreases to 20.60 at 284.89 Hz. In case of nickel oxide nanoparticles doped NLC, at 35 ℃ the value of $\boldsymbol{\varepsilon'}$is 28.21 at 31.74 Hz and decreases to 8.30 at 284.89 Hz. This shows that on doping of nickel oxide nanoparticles, there is no much effect on the $\boldsymbol{\varepsilon'}$ at high frequency but at low frequency it is highly affected. The $\boldsymbol{\varepsilon'}$ is found to be either constant or decreases as the frequency has been increased.[14,15,16] At higher frequency, the lower values of $\boldsymbol{\varepsilon'}$ propose that the molecules rotate about theirlong molecular axis.[17] At low frequency, the higher value of the $\boldsymbol{\varepsilon'}$ is due to effect of space charge polarization.[18] The decrease in $\boldsymbol{\varepsilon'}$ can be explained by Koop's theory.[19] The variation of $\boldsymbol{\varepsilon'}$ with frequency shows almost similar behavior for pure and nickel oxide doped nanoparticles NLC but the values of $\boldsymbol{\varepsilon'}$are found to be lower for doped NLC system.

When the nanoparticles are mixed in NLC system, the orientation of nanoparticles were in such a way that the dipole moment of nanoparticle is opposing the dipole moment of NLC causes to decreases the dielectric permittivity for doped system. The net dipole moment of doped system is decreased as compared to pure because the ionic polarizations are induced on the application of electric field.[20]

The variation of $\boldsymbol{\varepsilon}'$ with temperature is due to the crystal expansion, the electronic and ionic polarization also depends on purity (impurity) and perfection of crystals (crystal defect). At low temperature the variation is attributed to the crystal defect and ionic and electronic polarizations. At high temperature the variation is due to impurity dipoles and thermally generated charge carriers. The study of variation of $\boldsymbol{\varepsilon}'$ with temperature provides valuable information regarding the dipole reorientation of LC.[16,21,23] For pure NLC the maximum value of $\boldsymbol{\varepsilon}'$ has been found at 35℃. The decrease in $\boldsymbol{\varepsilon}'$ after 35℃ can be attributed to thermal agitation, where the randomization of dipoles takes place. Thus for pure nematic sample at transition temperature, the thermal agitation cause the anisotropic molecule to vary their original orientation. For the doped NLC the value of $\boldsymbol{\varepsilon}'$ increases considerably with increase in the temperature at the low frequency region and almost constant at high frequency. The contribution of space charge effect towards polarization may have tendency to increase with increase in the temperature.

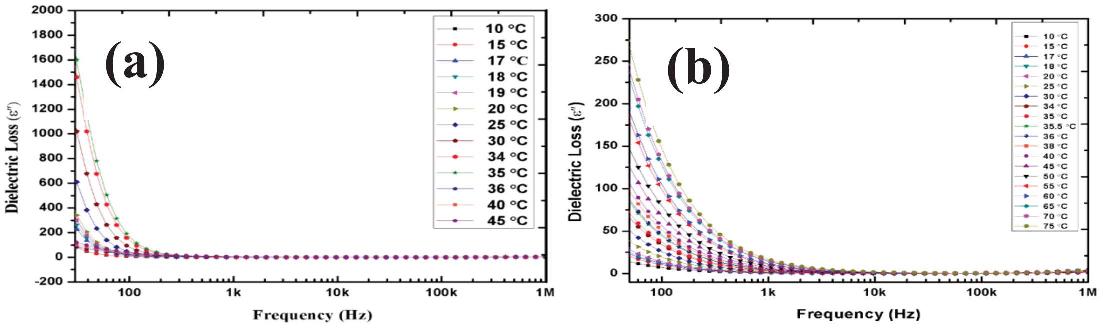


FIGURE 4. Behavior of dielectric loss ($\boldsymbol{\varepsilon}''$)as a function of log of frequency for (a) pure nematic liquid crystal (5CB) and (b) NiO nanoparticles doped nematic liquid crystal (5CB) at different temperatures.

Fig.4 shows the variation of dielectric loss with frequency for pure and NiO doped NLC at different temperatures. In case of pure and NiO nanoparticles NLC it is observed that the dielectric loss decreases with increasing frequency in low frequency region and shows almost constant behavior at high frequency region. The variation of dielectric loss with frequency shows almost similar behavior for pure and NiO nanoparticles doped NLC but the values of dielectric loss are lower for doped NLC. The low value of the dielectric loss in case of doped NLC as compared to pure NLC indicates good quality of the doped nanomaterials. At low frequency, the larger value of dielectric loss is due to space charge polarization owning to charge lattice defects.[22] At the low frequency region, the value of dielectric loss increaseswith increase in temperature and maximum value has been found at 35 ℃ and afterwards decreases the value of dielectric loss with increasing in the temperature for pure NLC. At high frequency the value of dielectric loss remains constant. In case of doped NLC the value of dielectric loss regularly increases with increase in the temperature at low frequency and remains constant value at high frequency.[23]

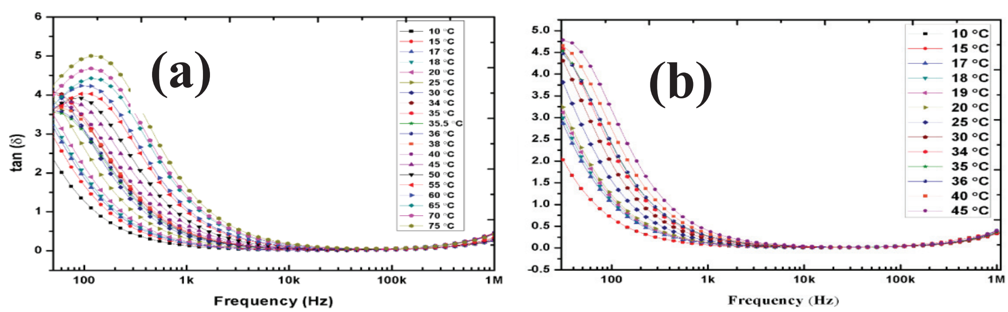


FIGURE 5. Behavior of dielectric loss factor (tan δ) as a function of log of frequency for (a) pure nematic liquid crystal (5CB) and (b) NiO nanoparticles doped nematic liquid crystal (5CB) at different temperatures.

Fig.5 shows the variation of dielectric loss factor (tan $\boldsymbol{\delta}$) with frequency for pure and NiO doped NLC at different temperatures. The tan $\boldsymbol{\delta}$ described the energy dissipation in the dielectric system. This is due to domain wall resonance. From the relation (4), tan $\boldsymbol{\delta}$ is directly proportional to the imaginary part of the complex dielectric constant, so it exhibits similar behavior. At high frequency the value of tan $\boldsymbol{\delta}$ are found to be low or almost constant for the pure NLC. The value of tan $\boldsymbol{\delta}$ is maximum at low frequncy region. The value of tan $\boldsymbol{\delta}$ decreses with increase in frequency. At 45℃, the value of tan $\boldsymbol{\delta}$ for pure NLC is 4.7 at 31.83Hz and decreases to 0.15 at 3302.4 kHz. This may be due to space charge polarization.[22] This indicates a peak for pure NLC. This low frequency peak is connected to the low frequency relaxations of the ions. This low frequency relaxation mode cannot be assigned any relaxation mode due to space charge accumulation near the substrate phase. The existance of relaxation is related to ionic impurity present in the material . The low frequency behaviour is due to the ionization-recombination assisted diffusion of slow ions in homogeneous aligned configuration for pure NLC.[20] In case of doped NLC the value of tan δ also decreases with increase in the frequency in low frequency region. At high frequency the values of tan δ are constant. At 45℃, the value of tan δ for doped NLC is 3.8 at 74.5Hz and decreases to 0.15 at 4026.6 kHz. For the doped case the peak is shifted downward indicates the impurity present in material. This due to fact that when nanoparticles are added to NLC, then some LC ions and nanoparticles ions cancel each other. The shifting of peak suggests that the relaxation frequency of the nanoparticles doped composite is different from that of pure sample and variation also depend upon the type of nanoparticles used and its compatibility with LC host.[17]

The values of tan δ are continuously increasing with rise in the temperature for pure NLC. In case of doped NLC the tan δ does not vary systematically with temperature. The tan δ increases with increase in temperature at 34 ℃ and at 35 ℃ it decreases with rise in temperature. At 36 ℃ it again increases in very small increment and value of tan δ is overlapping to previous value lie at low temperature. The variation of tan δ with frequency shows almost similar behavior for pure and NiOnanoparticles doped NLC but the values of tan δ are found to be lower for doped NLC.
It is also found that for pure NLC the tan δ has high values and it decreased when NLC is doped with NiO nanoparticles but it is gradually decreased in high frequency regime. The small peaks are absent in the high frequency region which suggests the relaxation processes or loss peaks. The peaking behavior arises when the migrating (hopping) frequency of the localized electric charge carrier approximately equals to that of frequency of the applied ac field.[24,29] The variation in the value of ε' and ε'' due to doping of NiO nanoparticles can be explained by considering the contribution of dipole moment of nanoparticles molecules in the host LC molecule. Each nanoparticles molecules have a preferential axis of orientation and it will aligned along the host LC molecule thereby resulting in the restriction on the freedom of its movement. Thus, the dipole moment of nanoparticles molecules contribute significantly to the actual value of ε' and ε'' and also relaxation frequencyvalue.[17,25]

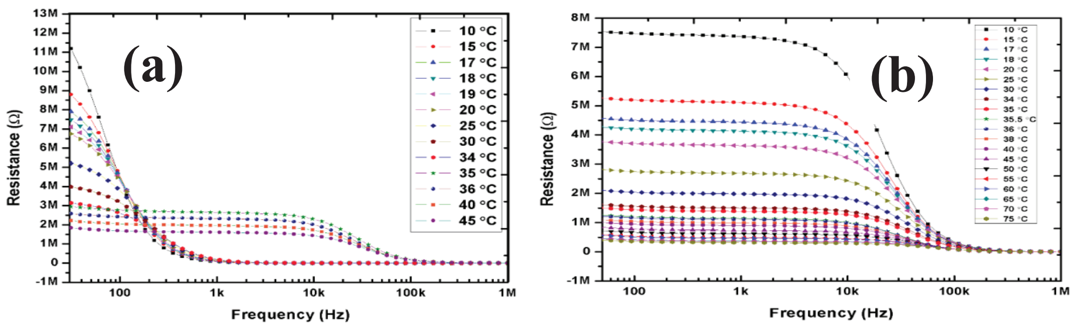


FIGURE 6. Behavior of resistance as a function of log of frequency for (a) pure nematic liquid crystal (5CB) and (b) NiO nanoparticles doped nematic liquid crystal (5CB) at different temperatures.

Fig. 6 shows the behavior of resistance of pure and NiO nanoparticles doped NLC material with frequency at different temperatures. The resistance-temperature behavior may be attributed to many factors such as occurrence of phase transition, presence of impurities and cations migration. The conductivity of NiO is directly connected to the number of electrons, electrons formed by the ionisation of nickel atom and the oxygen vacancies. The resistance of both pure and doped NLC decreases with rise in the temperature due to increase of preffered orientation of the molecules which is influenced by the temperature. The resistance of pure and doped NLC was decreased further rise the temperature. This is due to fact that with increasing the temperature the grain boundary and crystal lattice deficiencies of the LC is decreased, resulting in an increase of the mobility of the carriers. The LC director reorientation cause a change in resistance due to anisotropic of LC. The resistance has been reduced (or enhanced) on the doping NiO nanoparticles into NLC. The reason behind the occurrence of this low frequency relaxation mode lies in the relaxations of the ionic charges through single particle diffusion of fast ions and ionization–recombination-assisted diffusion of slow ions.[26]

Optical Textural Studies

The degree of light scattering in the absence of an applied field strongly depends on a number of factors such as the types of materials used, their composition, casting solvent and on the details of firm forming procedure. On the other hand, morphology of PDLC samples also influences the light transmittance properties in the presence of an applied field.[28] Optical textures are the patterns on liquid crystalline samples which are observed under microscopic, usually in polarized light. Change in texture at a particular temperature indicates the occurrence of phase transition. The morphology strongly depends upon the type of nanoparticles and LC used. In this part the microscopic texture of the pure and nickel oxide nano particles doped nematic liquid crystal has been discussed.[23]

The polarizing optical microscopic images of pure and NiO doped NLC system have been taken to ensure the proper dispersion of nanoparticles in the host material. A homogenous distribution of phase separated LC droplets can be clearly seen in both the composite films. The optical textures of pure and NiO doped NLC in a homogeneous cell under a crossed polarizer condition are shown in Fig. 7. Fig. 7(a) and 7(b) show the textures of the pure NLC in a homogeneous cell. Fig. 7(c) and 7(d) show the textures of NiO doped NLC in a planar cell.[27] With the transmitted light, the variation between the on and off states can be observed clearly. From these figures, it is seen that the molecular alignment of the NLC material is improved upon doping with NiO nanoparticles. The bright state (Fig. 7(d)) also shows an improved texture compared to pure NLC (Fig. 7(b)). This confirms the improvement of the contrast in NiO doped NLC system. Moreover, from the Fig. 7(c) and 7(d) it is clear that the NiO nanoparticles are uniformly dispersed in NLC and higher concentration of nanoparticles has led to formation of small aggregates of nanotubes in the NLC system.[27]

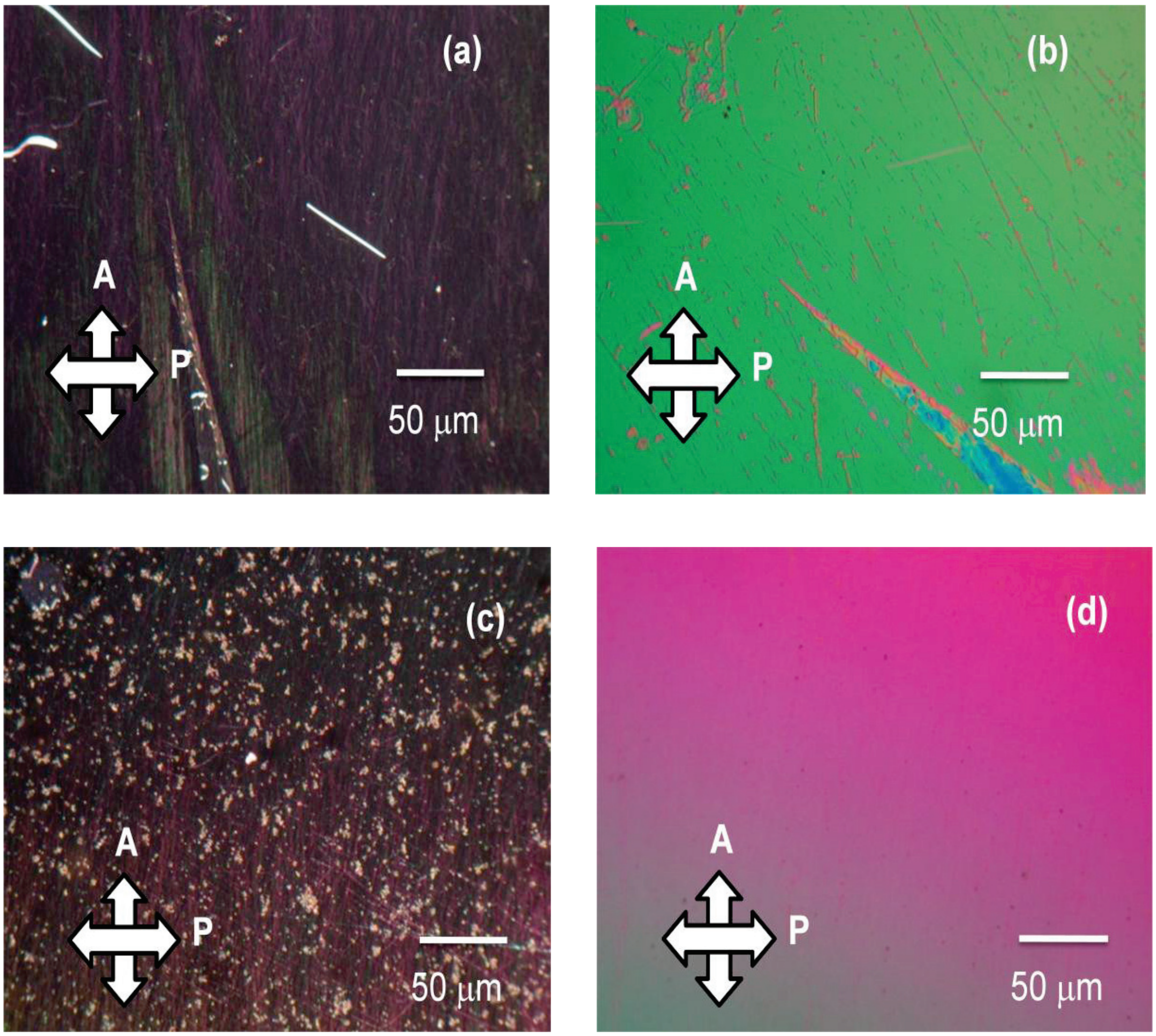


FIGURE 7. Polarizing optical micrographs of pure [(a) dark, (b) bright states] and NiO nanoparticles doped nematic liquid crystal cell [(c) dark, (d) bright states] at room temperature. Scale bar: 50 μm. Crossed arrows represent crossed polarizer (P) and analyzer (A).

CONCLUSIONS

The present paper demonstrates the improvement in the dielectric and optical parameters by doping of NiO nanoparticles in the NLC. It may be concluded that dielectric permittivity and dielectric loss values decreases with the addition of NiO nanoparticles. The NiO nanoparticles doping also decreases the value of tanδ which shows the low relaxation frequency of the ions. In low frequency region the Z has strong frequency dependence. The resistance has been reduced (or enhanced) on the doping of NiO nanoparticles into NLC. From optical textural investigation we concluded that the optical contrast has been improved NiO doped NLC.It is also clear that the NiO nanoparticles are uniformly dispersed in NLC and higher concentration of nanoparticles has led to formation of small aggregates of nanotubes in the NLC system.

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