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Original Research Article

Thermo-optical, dielectric, and structural investigation of a binary liquid crystal mixture doped with nanoparticles

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ABSTRACT

The unique electro-optical properties of liquid crystals (LCs) make them indispensable for modern displays and optoelectronic devices, with increasing interest in enhancing these properties via nanoparticle (NP) doping. In this work, we investigated the thermo-optical and electrical properties of a binary liquid crystal mixture doped with ferroelectric strontium titanate (SrTiO₃) nanoparticles at a concentration of 0.1 wt%. The suspension was synthesized using the ultrasonicator bath technique to ensure a homogeneous dispersion of the 100 nm nanoparticles. Characterization was performed through Differential Thermal Analysis (DTA), Refractive Index measurements, and Fabry-Perot Scattering Studies (FPSS) to determine the order parameter. Dielectric constant measurements were conducted to evaluate the electrical response, revealing that the incorporation of ferroelectric SrTiO₃ enhances the dielectric permittivity of the composite. Furthermore, Polarizing Optical Microscopy (POM) confirmed that the nanoparticles remained well-dispersed within the LC matrix without inducing macroscopic defects or disrupting the LC director. The resulting composite system demonstrates optimized properties, such as improved molecular ordering and enhanced dielectric response, which are not present in the pure LC mixture. These advancements hold significant potential for energy conservation applications, specifically in the development of high-efficiency smart windows and low-power optoelectronic displays.

1. Introduction

Liquid crystals (LCs) are a stable thermodynamic state of matter that exists between the solid and liquid phases. They exhibit a unique combination of properties from both: they can flow like a liquid but also possess some degree of molecular order, similar to a crystal. Despite their success, the demand for next-generation devices requires LCs with faster response times, lower power consumption, and higher thermal stability. Traditional chemical synthesis of new LC molecules is often time-consuming and costly. Consequently, 'doping' existing LC matrices with nanomaterials has emerged as a versatile and efficient strategy to tune their physical properties without altering their fundamental chemical structure. The incorporation of nanomaterials into liquid crystal (LC) systems creates a nanocomposite that exhibits properties distinct from its individual components. The nanomaterials act as dopants that can significantly alter the LC's properties, including its thermal, electro-optical, and dielectric behaviour. Electro-optical properties, which are critical for display applications, are heavily influenced by nanomaterial doping. The presence of nanomaterials can significantly enhance the phase stability of liquid crystalline phases, leading to a wider temperature range for the mesophase.

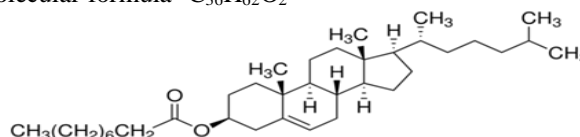
While single-component LCs are well-studied, binary LC mixtures are often preferred in industrial applications due to their wider mesophase temperature ranges and optimized visco-elastic properties. Investigating the interaction between a binary LC host and a low concentration (0.1 wt%) of SrTiO₃ nanoparticles provides a pathway to achieve high-performance composites that remain stable over time.

2. Materials

The main aim of this work is to characterize thermal and optical effects in mixtures of liquid crystal and their composite with ferroelectric nanoparticles. Such liquid crystal systems show strong thermo-optical properties.

A. Cholesteryl pelargonate (nonanoate)

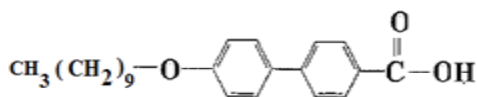
Molecular formula -C₃₆H₆₂O₂



B. 4' Decyloxybiphenyl -4 Carboxylic acid (D2041)



Molecular Formula –C₂₃H₃₀O₃



C. Ferroelectric Nanopowder- Strontium Titanate (SrTiO₃)

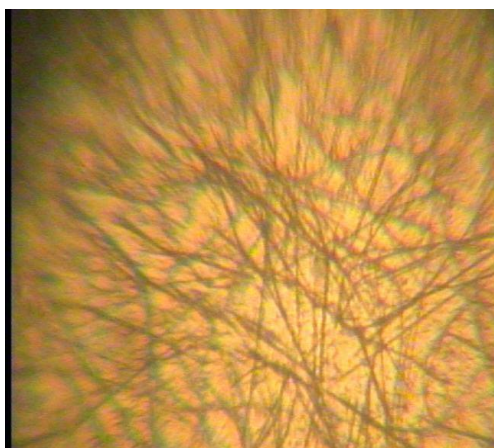
90 % wt. of A + 10 % wt of B + 0.1% wt of SrTiO₃

Doping a liquid crystal material is a rather straightforward technique. The two samples were first weighed accurately using analytical microbalance in the required proportions. The liquid crystals are mixed at room temperature and then the mixture is cycled above and below the highest clearing point, the point where the material is in an isotropic phase, for an hour to ensure good mixing.

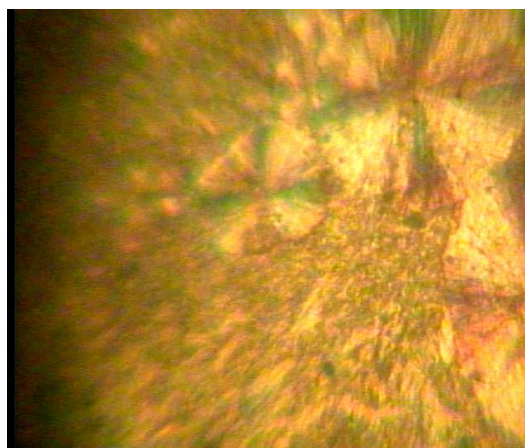
4. Methodology and discussion

4.1 POM study

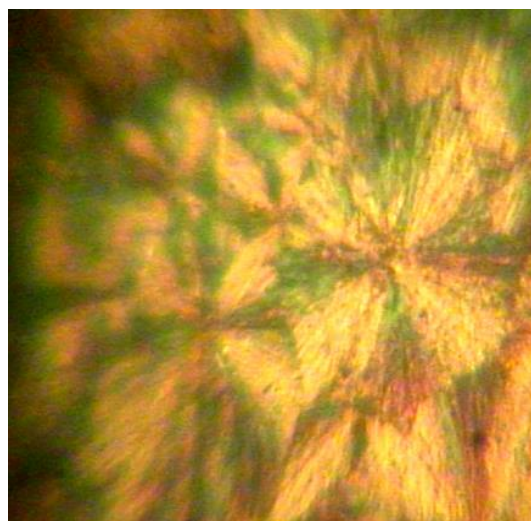
Textures of Binary mixtre



Nematic texture with surface disclination at 110 °C



Appearance of Sm A fan like textuer at 38°C



Crystalline phase at 34°C.

The thermal behavior and phase sequence of the pure binary liquid crystal mixture were characterized using Polarizing Optical Microscopy (POM). During the heating cycle, the sample undergoes a transition from the crystalline solid state to the liquid crystalline mesophase at 76°C reaching the isotropic liquid state (clearing point) at 115 °C. Upon cooling the sample from the isotropic melt, a Nematic (N)

phase emerges at 110°C, identified by the appearance of characteristic surface disclination lines. As the temperature is further reduced, a transition to the Smectic A (SmA) phase occurs at 38°C marked by the development of a classic focal conic fan-like texture. This SmA phase remains stable over a narrow temperature range of 4°C before the onset of the crystalline phase at 34°C.

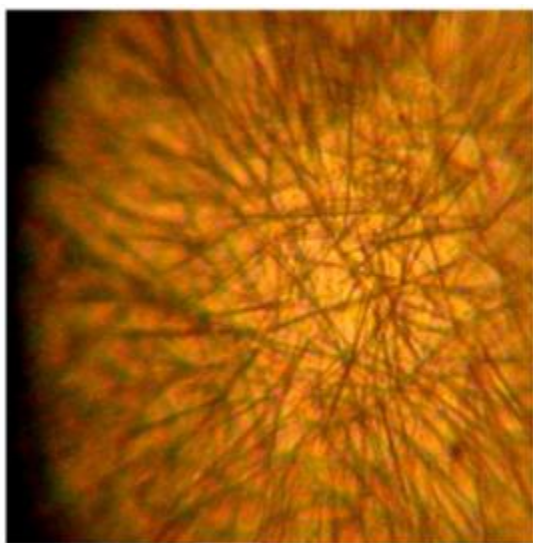
3. Objective of the study

This research aims to understand the interactions between nanoparticles and liquid crystals to improve the performance of existing technologies and enable new applications.

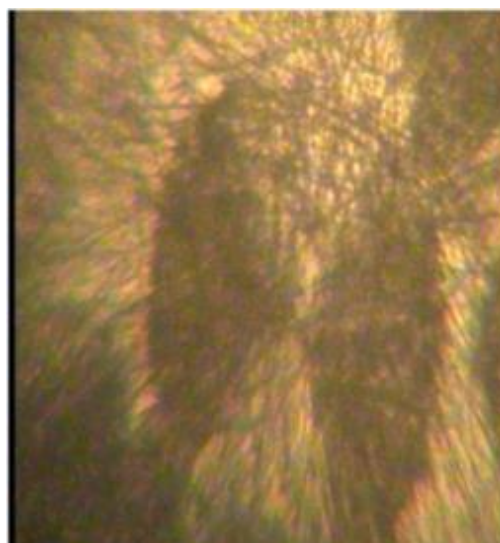
Doping LCs with nanoparticles allows researchers to tune and improve the material's electrical, thermo-optical properties characteristics.

The addition of nanoparticles can expand the temperature range over which the liquid crystal phase is stable, making devices more robust for a wider range of environmental conditions. The nanoparticles can alter the LC's refractive index which is one of the key parameters for light manipulation.

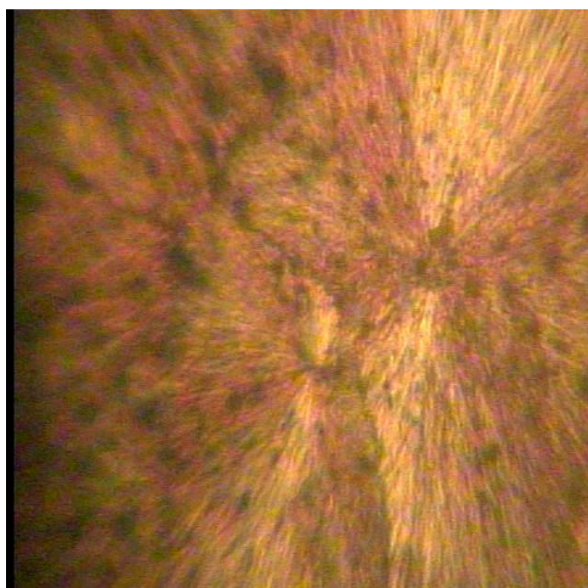
Textures of Binary mixture + 0.1% wt of SrTiO₃



Nematic texture with surface disclination at 123°C.



Appearance of Sm A fan like texture at 41°C.



Crystalline phase at 35°C.

The incorporation of 0.1 wt% SrTiO₃ nanoparticles into the binary liquid crystal mixture significantly altered its thermal stability and phase transition profile. Upon heating, the nanocomposite melts into the mesophase at 70°C, representing a 6°C reduction in melting temperature compared to the pure LC mixture. The isotropic transition temperature was observed to increase to 125°C, effectively extending the clearing point by 10°C.

During the cooling cycle, the Nematic (N) phase with characteristic surface disclination lines appeared at 123°C. Further cooling facilitated the transition to the Smectic A (SmA) phase at 41°C, identified by a well-defined focal conic fan texture. The sample eventually returned to the crystalline state at 35°C. Notably, the addition of ferroelectric nanoparticles expanded the Nematic phase stability by 10°C and the Smectic phase stability by 2°C. This dual effect lowered the melting point while elevating the isotropic

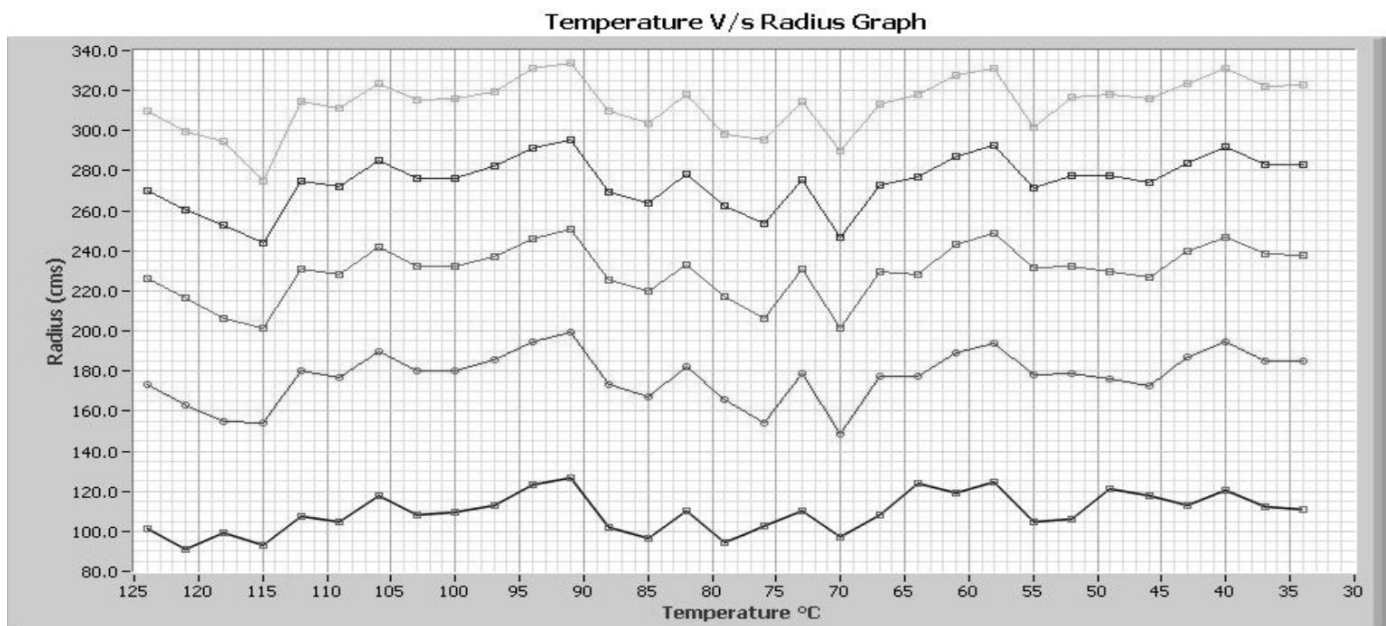
temperature results in a significantly broader operating temperature range for the liquid crystalline state.

4.2 Fabry perot scattering study

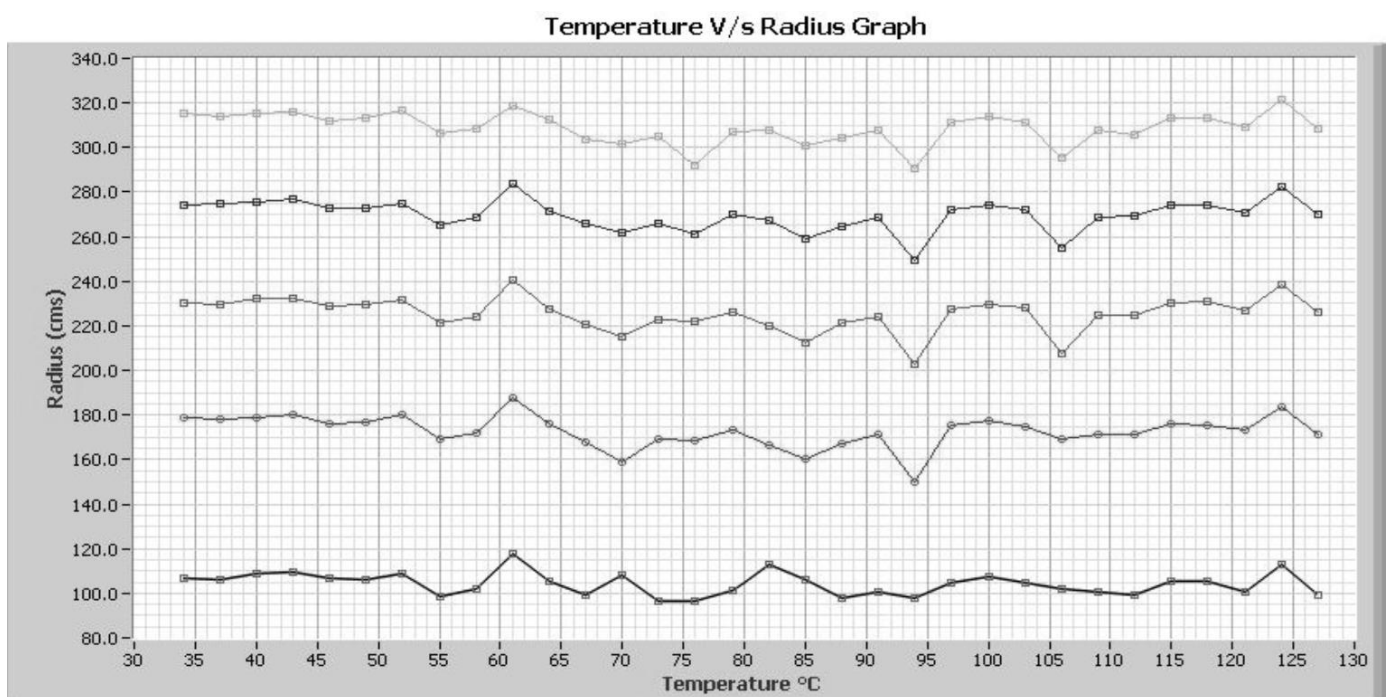
Phase transition temperatures for Cholesteric Nanoate and the its mixture with D2041 and doped mixture at various heating and cooling cycles were observed. Experiment is repeated for heating and cooling cycles. The optical technique of measuring the diameter of Fabry-Perot rings obtained from Fabry-Perot interferometer coupled with a spectrometer has been used to determine the phase transition temperatures of liquid crystal. This technique is highly sensitive enough to identify PTTs not detected earlier by other techniques.

The diameter of fabery-Perot rings are recorded as a function of temperature and plotted as shown in following figures:

(a) For Mixture



(b) For Mixture+ 0.1% concentration of Nano powder.

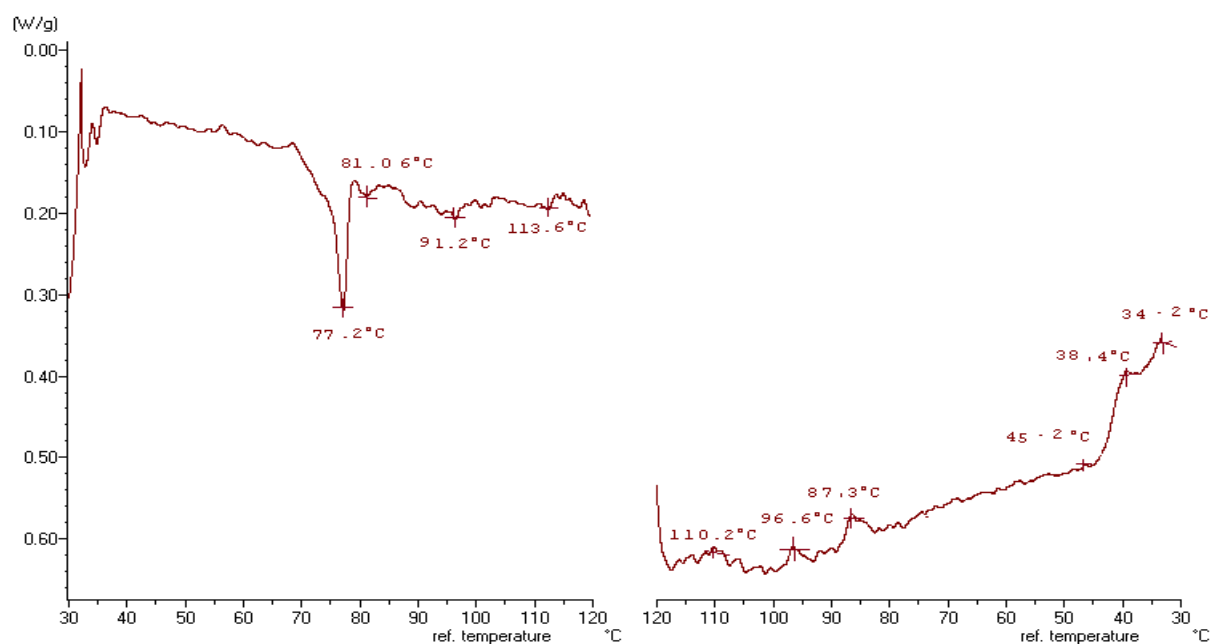


4.3 Differential thermal analysis

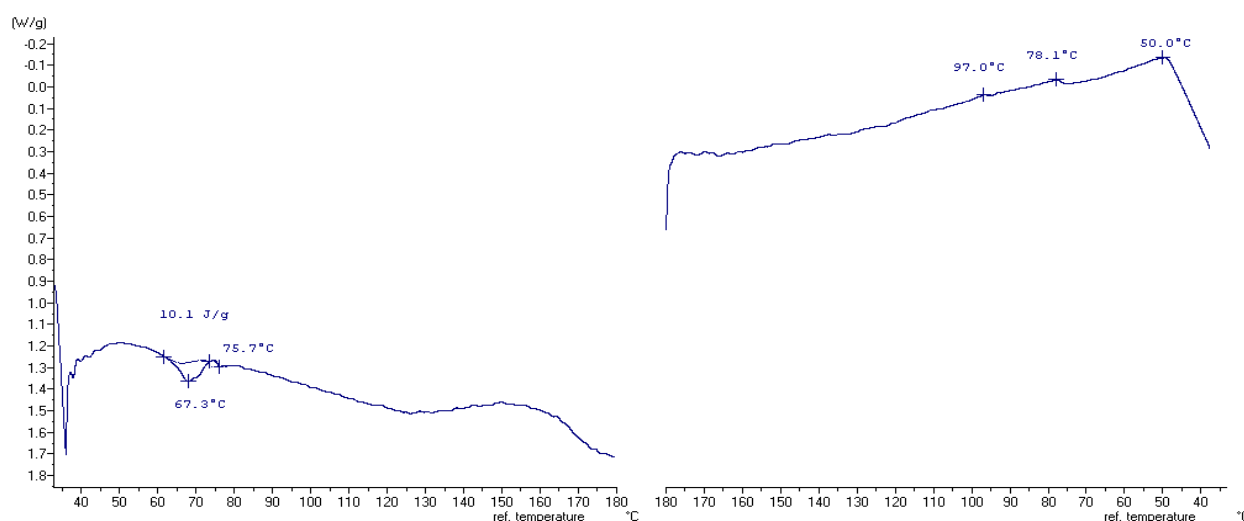
DTA involves heating or cooling a test sample and an inert reference under identical conditions, while recording any temperature difference between the sample and reference. This differential temperature is then plotted against temperature. The phase transition temperature of liquid crystals can be obtained from DTA curve. A balance between the sample and

the reference material is maintained by adding heat via the filament. When a sample melts, for example, from a crystalline solid to a SA phase, energy must be supplied to the sample to prevent an imbalance in temperature between the sample and the reference material. This energy is measured and recorded by the instrument as a peak on a baseline.

(a) For Mixture



(b) For Mixture+ 0.1% wt of nano powder.



Enthalpy Changes and Structural Order

The magnitude of the enthalpy change provides valuable information about the nature of the phase transition. The enthalpy change (ΔH) during a phase transition is a direct measure of the energy absorbed or released, and it is proportional to the change in structural ordering between the two phases. The melting of a crystalline solid to either a liquid crystal or an isotropic liquid involves a large enthalpy change, typically in the range of 30 to 50 kJ/mol. This is because the transition from a highly ordered, rigid solid lattice to a less ordered fluid state requires a significant amount of energy to

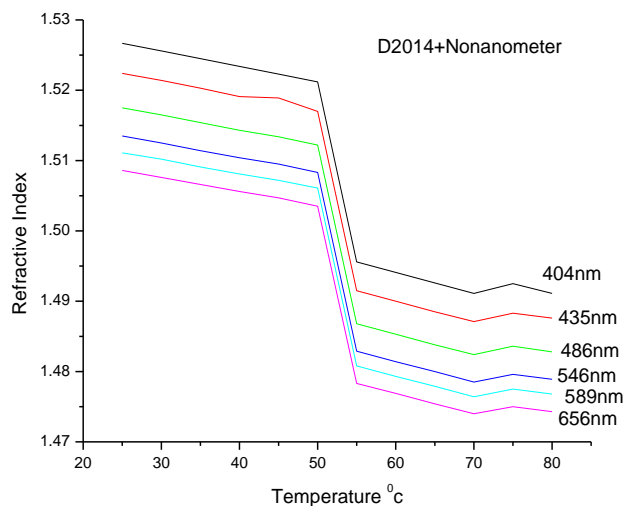
break the strong intermolecular bonds. Liquid Crystal to Liquid Crystal/Isotropic Liquid Transition: In contrast, the transitions between different liquid crystal phases (e.g., smectic to nematic) or from a liquid crystal to an isotropic liquid are associated with much smaller enthalpy changes. The liquid crystal (LC) to liquid crystal and LC-to-Isotropic Liquid transitions typically have enthalpy changes of around 4 to 6 kJ/mol. The small enthalpy changes in these transitions indicate that the molecules do not undergo a complete loss of order, as seen in the solid-to-liquid transition. Instead, there is a minor adjustment in positional order.

Table 1: Phase Transition Temperatures in °C by FPSS, DTA and POM techniques.

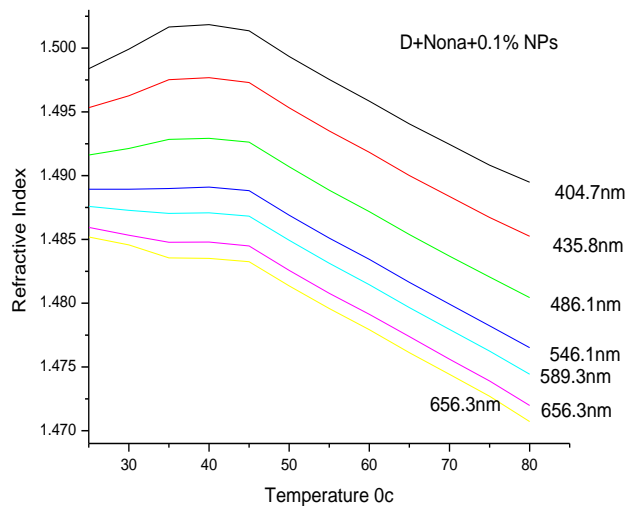
| Compounds | FPSS | DTA | POM |
|--|---|--|------------|
| Nonanoate+ D2041 | Heating-61,70,83,94,106,123 Cooling-115,106,89,83,73,56,40 | Heating- 77,81,91,113 Cooling- 105,96,87,45,38,34 | 34,38 ,110 |
| Nonanoate+ D2041+0.1% wt of SrTiO ₃ | Heating -49,67,85,103,110,125,136 Cooling-127,122,111,89,85,61,50,35 | Heating - 67, 75 Cooling - 97,78,50 | 35,41,123 |

4.4 Refractive index measurement

Refractive indices (RI) of the samples are measured using Abbe's refractometer at multiple wavelengths as shown in following figs. The accuracy of this refractometer is up to fourth decimal. For a given wavelength, we measured



ordinary refractive indices of all compounds with the intervals of 5°C. The refractive index decreases as the temperature increases. Near phase transition temperature it decreases rapidly which results in a density fluctuation. The RI of all the samples decreases with increase in wavelengths.



4.5 Refractive index as a function of temperature

The refractive index (RI) of the D2014 + Nonanoate liquid crystal mixture was investigated as a function of temperature and for mixtures with 0.1% wt Nano powder.

Behavior of Pure D2014 + Nonanoate

The pure D2014 + Nonanoate mixture exhibits a continuous decrease in refractive index with increasing temperature. This is a typical behaviour for liquid crystals as increasing temperature reduces the orientation order, leading to a decrease in the material's anisotropy.

1. From an initial temperature below 50°C, the RI gradually decreases.
2. A notable, sharp decrease in RI occurs in the temperature range of 50°C to 55°C.
3. The RI continues to decrease, but more gradually, from 55°C up to 70°C.
4. An interesting and uncharacteristic increase in RI is observed at 75°C, after which it again decreases at 80°C as the material transitions to the isotropic state.

Behavior of Doped Mixtures

The addition of nonanoate nanoparticles at concentrations of 0.1% wt significantly alters the temperature-dependent RI behavior, particularly at lower temperatures.

1. The RI initially increases in the range of 35°C to 40°C. This is an unusual effect that is not seen in the pure mixture and may indicate a change in molecular alignment induced by the nanoparticles. Following this increase, the RI remains constant up to 50°C.
2. From 50°C to 70°C, the RI decreases, mirroring the general trend of the pure sample.
3. Similar to the pure sample, an increase in RI is observed at 75°C, followed by a final decrease as the mixture approaches the isotropic state at 80°C.

The observed differences in the refractive indices of your liquid crystal (LC) mixture and doped mixtures, along with their temperature-dependent behavior, are caused by changes in molecular structure and arrangement. The addition of nanoparticles introduces new interactions that modify the LC's properties

4.6 Order parameter

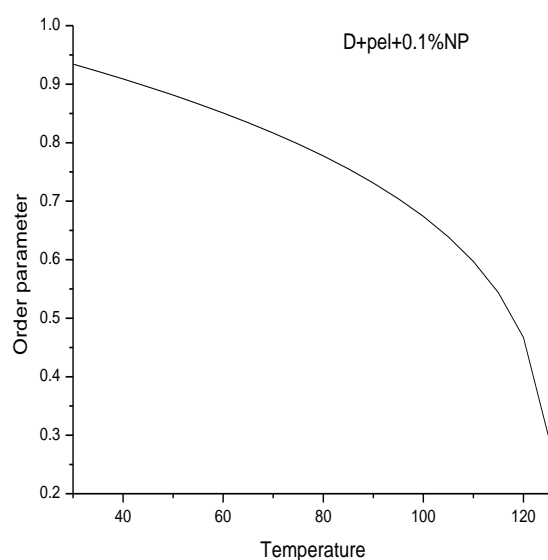
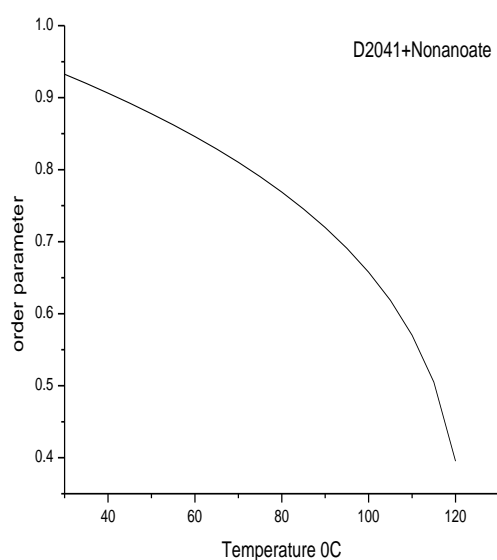
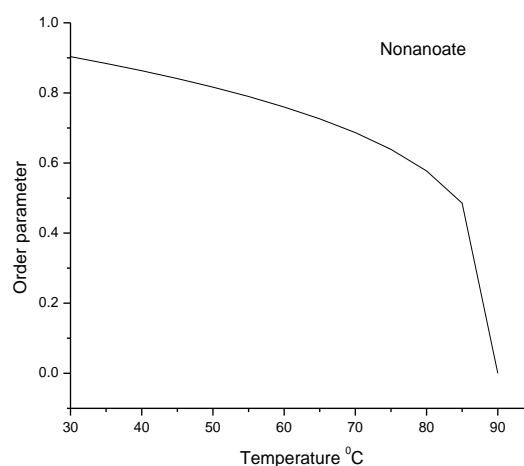
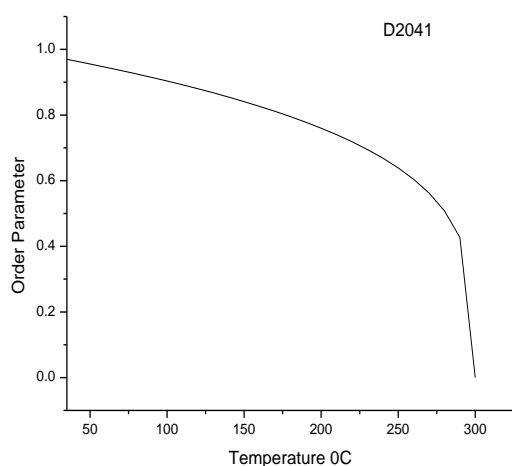
The order parameter is one of the most important parameters in the characterization of LCs, which can be determined in both microscopic and macroscopic forms. This parameter controls all physical properties of LCs in addition to verifying their degree of intrinsic long-range order

Order Parameters of all the samples are estimated from Haller's equation.

$$S = \left(\frac{T_c - T}{T_c} \right)^\beta$$

where T_c is clearing temperature and β is constant, depends on the molecular structure and its value is close to 0.25.

The variation of the order parameter with temperature and concentration is fundamentally dependent on the orientational order of the molecules. As temperature rises, thermal energy increases, causing greater molecular motion and a loss of alignment, which is reflected in a continuous decrease in the order parameter until it drops to zero at the nematic-to-isotropic transition temperature.



The LC mixture containing Ch. Nonanoate has an order parameter that is lower than that of the pure LCs but higher than that of Ch. Nonanoate alone. This is because the mesogenic dopants disrupt the host LC's orientational ordering more effectively than the host molecules can interact with themselves, leading to a net decrease in overall order. The addition of nanoparticles (NPs) to the LC mixture leads to an increase in the order parameter with increasing NP concentration. This suggests that the nanoparticles do not disrupt the LC's order in the same way as the mesogenic dopant.

At a low concentration of 0.1%wt NPs, the difference in the order parameter is minimal compared to the pure LC mixture. This indicates that the initial influence of the nanoparticles is to enhance the molecular interactions within the LC host, promoting a more stable orientational arrangement.

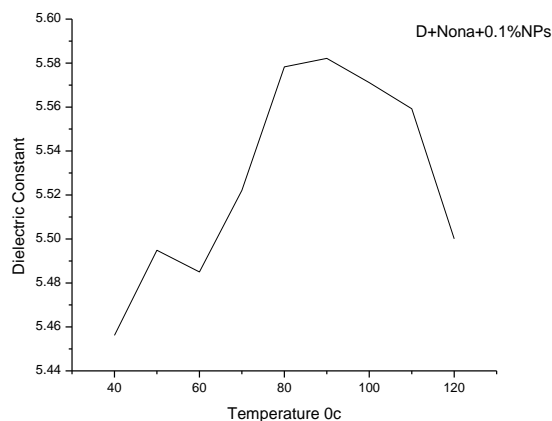
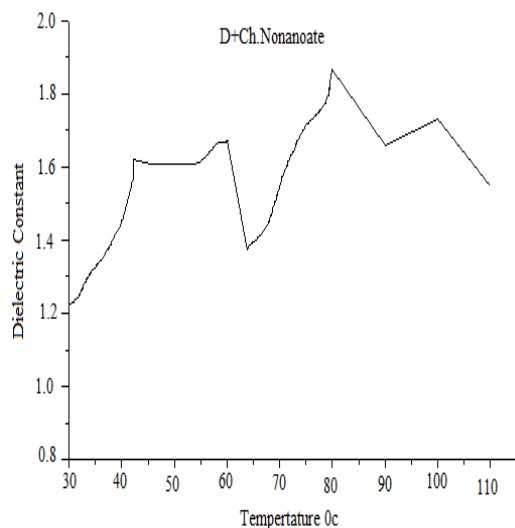
4.7 Dielectric properties

Measurement Dielectric Constant (ϵ') Dielectric constant is a physical quantity that describes how an electric field affects in dielectric medium and is determined by the

ability of a material to be polarized in response to an applied electric field inside the material.

Effect of Temperature

A significant enhancement in the dielectric permittivity was observed upon the incorporation of 0.1 wt% SrTiO₃ nanoparticles into the binary LC mixture. The dielectric constant increased from 1.23 for the pure LC mixture to 5.40 for the SrTiO₃-doped composite. This substantial shift indicates a dramatic increase in the net polarizability of the composite system. This four-fold increase in the dielectric response indicates a substantial improvement in the material's polarizability and its sensitivity to external electric fields. Strontium Titanate is a ferroelectric material. Unlike standard particles, ferroelectric NPs have a high intrinsic dielectric constant and a permanent dipole moment. Charges can accumulate at the interface of the nanoparticle and the liquid crystal. This creates interfacial polarization, which adds a further boost to the measured dielectric constant, especially at lower frequencies.



5. Result

In this paper it is reported that low concentrations of ferroelectric nanoparticles dispersed into the LC mixtures behave as a molecular dipole and can increase the clearing temperature. Without disrupting the structure and composition of the LC mixture, SrTiO₃ shares its intrinsic features with them by enhancing the thermos optical properties of LC mixture. The composite with nanoparticles have the capacity to exhibit improved properties like enhanced optical, electrical and mechanical properties. Doped LCs has wide range of applications such as flat, ultra-high definition, smart displays, optical polarizer etc. A study of temperature dependence helps in formulating liquid crystal mixtures that maintain stable performance over the wide operating temperature ranges required for devices like televisions, laptops, and automotive displays. Doping LCs with nanoparticles can make these displays more energy-efficient.

The observed increase in the order parameter with nanoparticle doping is a significant finding. This behavior can be attributed to the specific interactions between the nanoparticles and the LC molecules. Nanoparticles can act as nucleation sites for orientational ordering, effectively promoting the alignment of surrounding LC molecules and thereby increasing the overall order of the system. This molecular-scale structural rearrangement to a lower-energy, more ordered state is a key outcome of the nanoparticle-LC composite system, which is crucial for enhancing the performance of LC-based devices. The observed increase in the dielectric constant is highly beneficial for optoelectronic applications.

Authors' contributions

The author reviewed and approved the final version of the manuscript for publication.

Conflicts of interest

The author declares no conflict of interest.

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Data availability

No new data were created.

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