A comparative study of the thermodynamical parameters of cholesteric liquid crystal mixtures

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Abstract

The work presents a comparative analysis of the acoustical and thermodynamical aspects of Cholesteric Liquid Crystal (CLC) solutions. Pure CLC and polymer-dispersed Cholesteric Liquid Crystal (PDCLC) are the solutions under investigation. The PDCLC combination is prepared using a polymerization induced phase separation process. The optical polarizing microscope (OPM) and ultrasonic interferometer were used to examine CLC solutions. Ultrasonic velocity of the samples is employed to estimate a number of physical characteristics at different temperatures, including viscosity, acoustic impedance, relative association, surface tension, intermolecular free path length, ultrasonic attenuation, adiabatic compressibility, Wada constant, Rao's constant, and relaxation time of mixtures.

Keywords: Polymer-dispersed Cholesteric Liquid Crystal, Cholesteric Liquid Crystal, Polymer, Ultrasonic interferometer, Optical polarizing microscope, Clearing temperature

Introduction:

Mesophases with properties halfway between those of liquid and anisotropic solids are called liquid crystals (LC). The anisotropic nature of crystalline materials and the fluidity of the liquid state are demonstrated by the optical and physical (acoustic, mechanical, electrical and thermal) characteristics of LCs. With helical molecule architectures and temperature-dependent phase transitions, cholesteric LCs are thermotropic LCs. The LC molecules in CLCs self-organize into layers without regard to spatial ordering. The layers form a helix as the director changes periodically. Pitch of LC is the distance needed to execute one director revolution. Selective reflection, often known as Bragg's scattering for visible light wavelengths, is caused by the physical characteristics of short-pitch CLC [1,2]. Extrinsic perturbations that affect the length and

twisting direction of the helical pitch of CLCs include electric fields, magnetic fields, dyes, solvents, polymers and nanoparticles. By adding the appropriate proportions of dopants (dyes, polymers and nanoparticles), it is easy to change the pitch of the CLC helix [3-5], thereby changing the selective reflection.

After pure CLC is doped with exogenous substances, researchers have observed changes in its characteristics. The altered characteristics are used in sensors, optical fibers, and optoelectronic devices. For use in industries, technology, material science, and displays, researchers are extensively examining the characteristic features of both pure and doped CLC [3-12]. Our study focuses on analysing the acoustical and thermodynamical properties of CLC and polymer-dispersed CLC mixtures.

The preparation of PDCLC involves the addition of LC material in-to polymer matrix, giving it the flexibility of soft liquid-crystal materials. This substance is widely used in light shutters, smart windows, as well as in display technology. Due to their electronically regulated switching abilities, PDLCs have garnered significant attention in the scientific community. While there are multiple methods to prepare PDLC, the phase separation approach is the most straightforward.

The phase separation process can be used to produce liquid crystal droplets with a constant size and morphology, improving its electro-optical characteristics to be utilized in industry and technology. The assembly of LC droplets in polymer matrix affects the characteristics of polymer-dispersed LC. Thermal, polymerization, and solvent procedures are among the phase separation techniques. Of them, the simplest approach for producing polymer-dispersed liquid crystal systems is polymerization induced phase separation procedure. In this process, a prepolymer is added with liquid crystal (low molecular weight) to form a uniform mixture of polymer-dispersed LC material. Organic materials known as PDLCs are used in display technology and electro-optical systems [4,13-14,15-19].

In their investigation of dye and polymer doped liquid crystal composites, the inclusion of the dopant materials altered the isotropic temperature, texturing, and phase transition temperatures of pure liquid crystal according to researchers S.J. Gupta et al. [20]. Kanwar et al. [21] investigated the properties of PDCLC composites and discovered that the percentage transmittance of these composites is influenced by the amount of LC present in the polymer. Rita et al. [22] revealed

that the refractive indices of PDCLC mixture altered, which could serve as a conduit for the signal. M. Pandey et al. [23] found that once the polymer was dispersed in liquid crystal, the LC's birefringence was increased. Incorporating PDLC into various material applications necessitates a thorough examination of its morphology, transition temperatures, and both physical and optical characteristics.

The creation of LC composites, which can alter the characteristics of LC and add more utility to the material, has been focus of increased research recently. The advancement of next-generation displays, smart windows, sensors, and wearables is expected to be advanced and expanded by composite films made by PDCLC materials. which offer the advantages of ease of preparation, affordability, large-area manufacturing, and flexibility [12-13,17-18].

The acoustical characteristics of a polymer-dispersed cholesteric LC are compared with that of a pure cholesteric liquid crystal and are presented in this chapter's acoustical investigation. Pure cholesteric liquid crystal is added to polymer to create a PDCLC combination via a phase separation technique called polymerization induced phase separation. An optical polarizing microscope and an ultrasonic interferometer are used to characterize the liquid crystal composite. Using an ultrasonic interferometer to measure acoustical velocity at different temperatures yields the acoustical parameters. The samples' ultrasonic velocity is used to evaluate the different thermodynamic characteristics. An optical polarizing microscope is used to determine the liquid crystal samples clearing temperature.

Materials

Sigma Aldrich is the source of all liquid crystals, solvents, and polymers. Toluene rectified LR with a 99% purity and a synthesis grade is the achiral solvent utilized for the liquid crystal dissolution. Methyl methacrylate, a monomer with a 99.9% purity, is the polymer utilized. Every chemical was utilized just as it was delivered, requiring no additional purification.

Cholesteryl Pelargonate(97% purity) is the cholesteric liquid crystal used.

Methyl methacrylate is the polymer.

Toluene is the solvent.

The following lists the specifications for the different materials and solvents utilized.

Cholesteryl Pelargonate(CP):

Another name for it is cholesteryl nonanoate;

Its formula is $C_{36}H_{62}O_2$.

Weight in molecules is 526.88 g/mol

Methyl methacrylate:

Also known as MMA, methyl methacrylate is a monomer having

$C_5H_8O_2$ is the molecular formula.

Weight in molecules: 100.12 g/mol

101 °C is the boiling point.

940 kg/m³ is the density.

Toluene:

The solvent is colourless and has

Formula for molecules: C_7H_8

Weight in molecules: 92.14 g/mol

Density: 867 kg/m³

110.6 °C is the boiling point.

Preparation of PDCLC:

Phase separation is the technique employed to produce PDCLC films. The most effective method for producing polymer-dispersed liquid crystals among the different phase separation approaches is the PIPS (polymerization induced phase separation) approach. We prepared the PDCLC for our investigation using the PIPS approach. When the CLC is mixed with the monomer MMA, it entirely dissolves in the MMA at room temperature.

Samples S1: Pure CP

S2: 4%wt CP in MMA

Methodology: An optical polarizing microscope and ultrasonic interferometer are employed to analyse the cholesteric liquid crystal mixtures.

Optical Polarizing Microscopy (OPM):

With the use of an optical polarizing microscope coupled with a hot stage, remote sensing infrared temperature sensor, and camera, the clearing temperature of the CLC samples is determined. The temperature changes for the CLC samples are obtained using a heating coil that was designed locally. The temperature was measured using a non-contact infrared digital thermometer that was accurate to one decimal place.

Ultrasonic interferometer:

By determining the wavelength and at known frequency, an ultrasonic interferometer may precisely measure the sound velocity in a fluid (water). The measurement cell and the frequency generator (multi-frequency) are the different components of the ultrasonic interferometer. To keep the sample liquid at a known, steady temperature, the measurement cell's double wall permits water to circulate around it. An important source of information regarding the molecular nature and extent of intermolecular forces in liquid crystal samples is a crucial source of information about the propagation of sound waves through these mixtures.

The characteristics of liquid crystal intermolecular forces in liquid crystals affects their physical and optical characteristics, which undergo dramatic changes at PTT's (phase transition temperatures). In our work, we use an ultrasonic interferometer set to 1MHz at and close to PTTs to record sound wave velocity in liquid crystal samples. The water bath's constant temperature was used to achieve the temperature variation. Various physical parameters are evaluated using the ultrasonic velocity of LC samples at different temperatures, including ultrasonic attenuation, acoustic impedance, viscosity, relative association, surface tension, intermolecular free path length, adiabatic compressibility, Wada constant, Rao's constant, and relaxation time of mixtures. These thermodynamic variables determine the kind of intermolecular forces and structural changes occurring in the CLC solutions [24-32]. We can use these combinations in multiple ways if we know the precise variation.

Acoustical and thermodynamical variables of cholesteric liquid crystal samples:

1. Ultrasonic velocity (v) [26]

   $v = \lambda f = 2df$

where, λ - wavelength of sound wave and λ = 2d

d is the separation between neighbouring anode current peaks.

f is the sound wave frequency and is kept constant at 1 megahertz.

2. Surface Tension (σ) [27]

$$v = \left(\frac{\sigma}{6.3x10^{-4}\rho}\right)^{2/3}$$

where ρ is the CLC sample's density.

3. Viscosity (ŋ) [27]

$$\frac{\eta}{\sigma} = \frac{16}{15}\sqrt{\frac{M}{RT}}$$

where M is the sample's molecular mass,

and T is its temperature (K)

4. Acoustic Impedance (Z) [28]

$$Z = v\,\rho$$

5. Adiabatic compressibility (β) [28]

$$\beta = \frac{1}{\rho v^2}$$

6. Intramolecular free path length (L_f) [29]

$$L_f = K\ \beta^{\frac{1}{2}}$$

where *K* is Jacobson's constant that depends on temperature.

7. Ultrasonic attenuation ($\frac{\alpha}{f^2}$) [29]

$$\frac{\alpha}{f^2} = \frac{8}{\rho}\frac{\eta\pi^2}{v^3}$$

8. Relaxation time (τ) [29]

$$\tau = \frac{4\eta}{3\rho v^2}$$

9. Relative association (R A) [29]

$$RA = \left(\frac{\rho}{\rho_0}\right)\left(\frac{v_0}{v}\right)$$

where, ρ_0 and ρ are the density of Toluene & CLC sample respectively

v_0 and v are ultrasonic velocity in Toluene and in CLC sample respectively

10. Molar compressibility (Wada Constant) (W) [30]

$$W = \frac{M}{\rho}\beta^{\frac{-1}{7}}$$

11. Molar sound velocity (Rao's constant) (R) [30]

$$R = \frac{M}{\rho}\ v^{\frac{1}{3}}$$

where

M=M1W1+M2W2

(M1and M2 are the molecular weight of solute and solvent in CLC solution respectively)

(W1and W2 are the weight fraction of solute & solvent in CLC solution respectively)

Observation:

The clearing temperature for Cholesteryl Pelargonate and PDCLC, as obtained with optical polarising microscope, is 90^0C and 70^0C respectively. A variety of acoustical and thermodynamic parameters can be evaluated by measuring the ultrasonic velocity of cholesteric liquid crystal solutions for temperatures that vary from the room temperature up to 90^0 C. Because of the limitations of the measuring device, we have restricted our observation to 90^0C.

Table 1 shows the sound velocity and associated acoustical and thermodynamic parameters of the mixture of cholesteryl pelargonate and MMA.

Table 1 Acoustical parameters of a mixture of PDCLC composite

Temp (C)	**Velocity m/s**	**ŋ (10^{-4})**	**β (10^{-10})**	L_f **(10^{-11})**	**Z (10^5)**	**RA**	$\frac{\alpha}{f^2}$ **(10^{-14})**	**τ (10^{-13})**	**R (10^{-3})**	**W (10^{-3})**
30	1177	4.05	7.55	5.64	11.25	0.90	2.05	4.07	0.95	1.81
35	1157	3.91	7.81	5.74	11.06	0.90	2.09	4.08	0.95	1.81
40	1137	3.78	8.09	5.84	10.87	0.90	2.12	4.08	0.94	1.80
45	1116	3.65	8.40	5.95	10.67	0.90	2.17	4.08	0.94	1.79
50	1097	3.53	8.69	6.05	10.49	0.90	2.21	4.09	0.93	1.78
55	1080	3.42	8.97	6.15	10.33	0.90	2.24	4.09	0.93	1.77
60	1057	3.29	9.36	6.28	10.11	0.90	2.30	4.10	0.92	1.76
65	1034	3.16	9.78	6.42	9.89	0.90	2.36	4.11	0.91	1.75
70	1031	3.12	9.83	6.44	9.86	0.89	2.35	4.09	0.91	1.75
75	1017	3.03	10.11	6.53	9.72	0.88	2.38	4.09	0.91	1.74
80	977	2.84	10.96	6.80	9.34	0.90	2.51	4.14	0.90	1.72
85	960	2.74	11.35	6.92	9.18	0.90	2.56	4.15	0.89	1.71
90	944	2.66	11.73	7.03	9.03	0.89	2.61	4.16	0.89	1.70

Table 2 provides the sound velocity and associated acoustical and thermodynamic variables of the mixture of cholesteric liquid crystal (CP) and toluene (T).

Table 2 Acoustical parameters of a mixture of CP in T										
Temp (C)	**Velocity m/s**	**ŋ (10^{-4})**	**β (10^{-10})**	**L_f (10^{-11})**	**Z (10^5)**	**RA**	**$\frac{\alpha}{f^2}$ (10^{-14})**	**τ (10^{-13})**	**R (10^{-3})**	**W (10^{-3})**
30	1288	4.22	6.96	5.42	11.16	0.99	1.80	3.91	1.16	2.17
35	1268	4.09	7.18	5.50	10.98	0.99	1.83	3.91	1.15	2.16
40	1245	3.94	7.45	5.60	10.78	0.99	1.86	3.92	1.15	2.15
45	1232	3.85	7.61	5.66	10.67	0.99	1.88	3.91	1.14	2.14
50	1206	3.70	7.94	5.79	10.45	0.99	1.92	3.92	1.14	2.13
55	1188	3.59	8.18	5.87	10.29	0.99	1.95	3.92	1.13	2.12
60	1169	3.48	8.45	5.97	10.13	0.99	1.98	3.92	1.12	2.11
65	1154	3.39	8.66	6.04	10.00	0.98	2.01	3.91	1.12	2.10
70	1126	3.24	9.11	6.20	9.75	0.99	2.07	3.93	1.11	2.09
75	1106	3.13	9.44	6.31	9.58	0.99	2.11	3.94	1.10	2.08
80	1091	3.05	9.70	6.40	9.45	0.99	2.14	3.94	1.10	2.07
85	1083	2.99	9.85	6.44	9.38	0.98	2.15	3.93	1.10	2.06
90	1046	2.82	10.55	6.67	9.06	0.99	2.25	3.97	1.08	2.04

Results:

Figures 1 through 9 illustrate the variation of following variables: ultrasonic velocity, adiabatic compressibility, relative association, acoustic impedance, relaxation time, Wada constant, Rao's constant, ultrasonic attenuation, and intermolecular free path length of LC and polymer dispersed CLC.

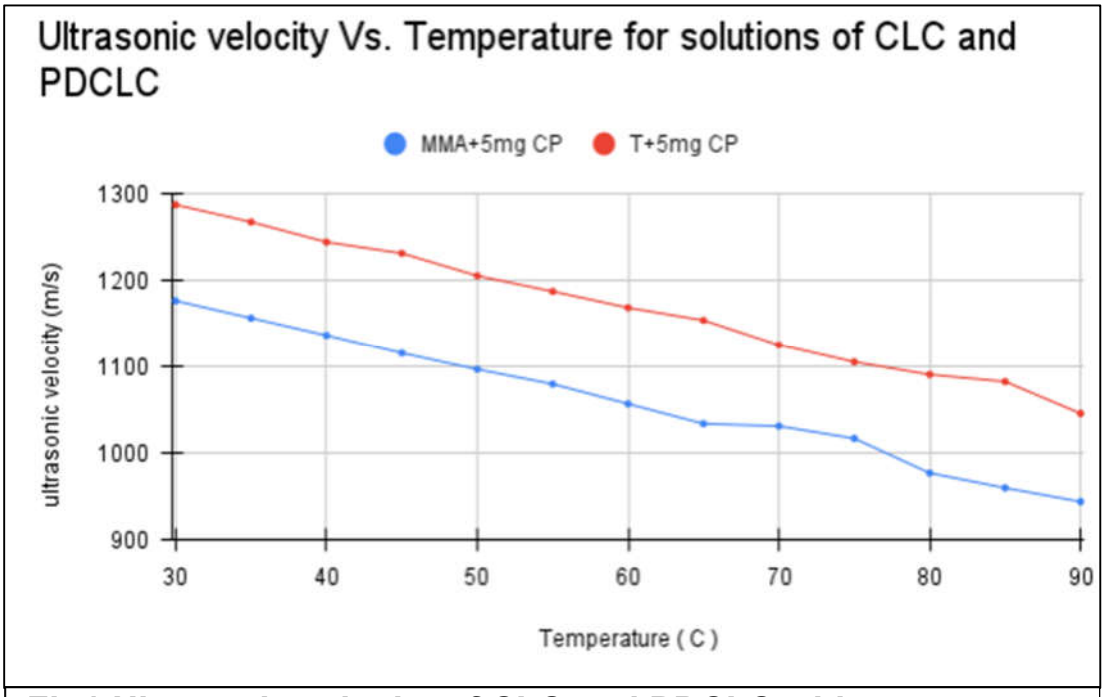

Fig1 Ultrasonic velocity of CLC and PDCLC with temperature

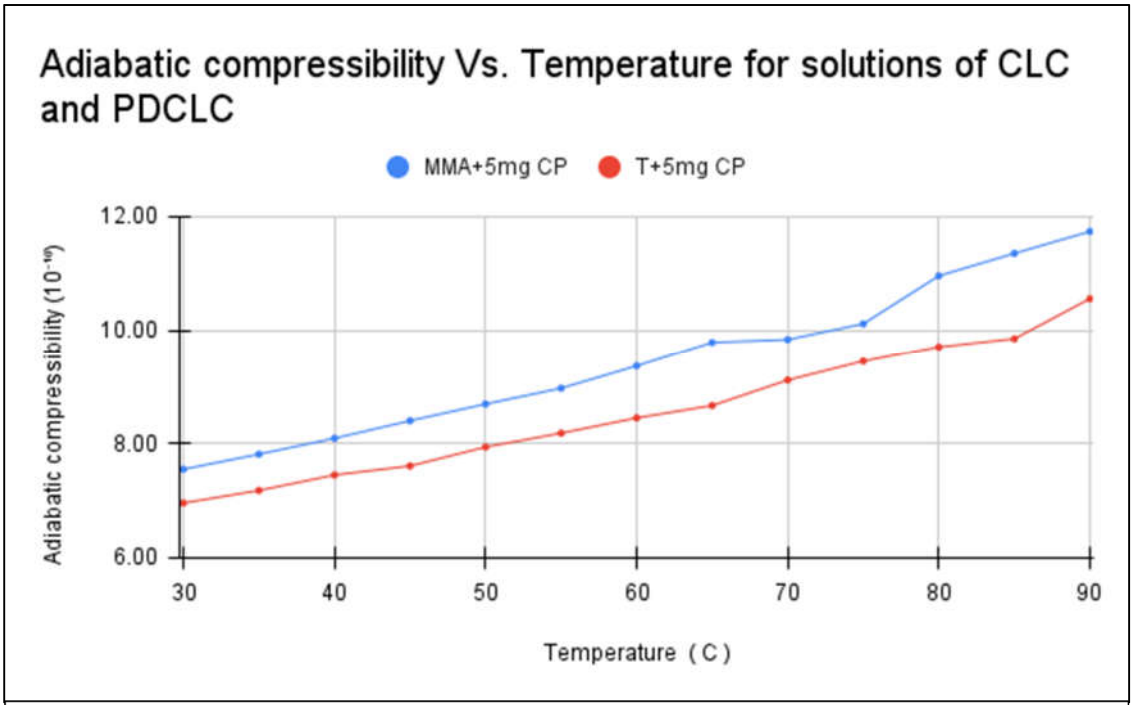

Fig2 Adiabatic compressibility of CLC and PDCLC with temperature

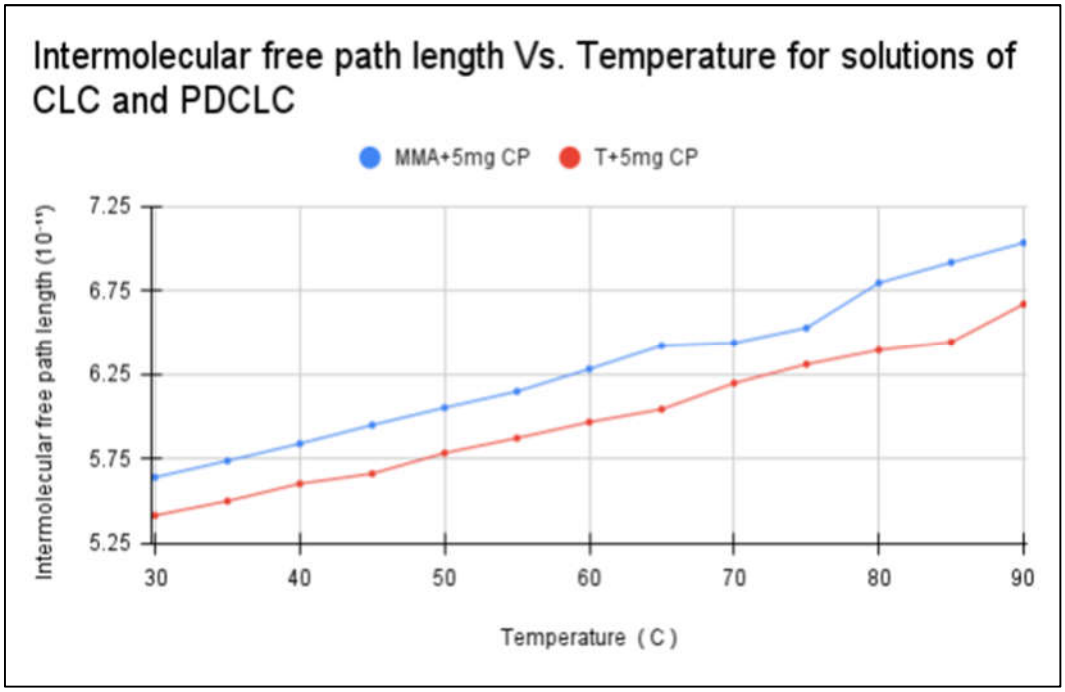

Fig3 Intermolecular free path length of CLC and PDCLC with temperature

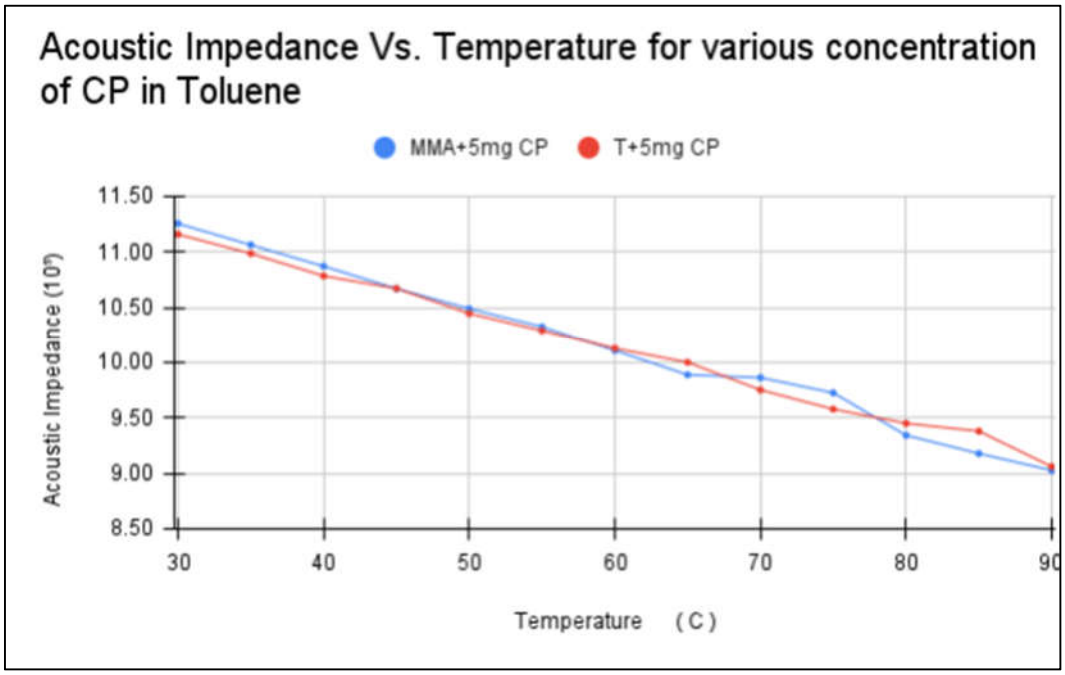

Fig4 Acoustic impedance of CLC and PDCLC with temperature

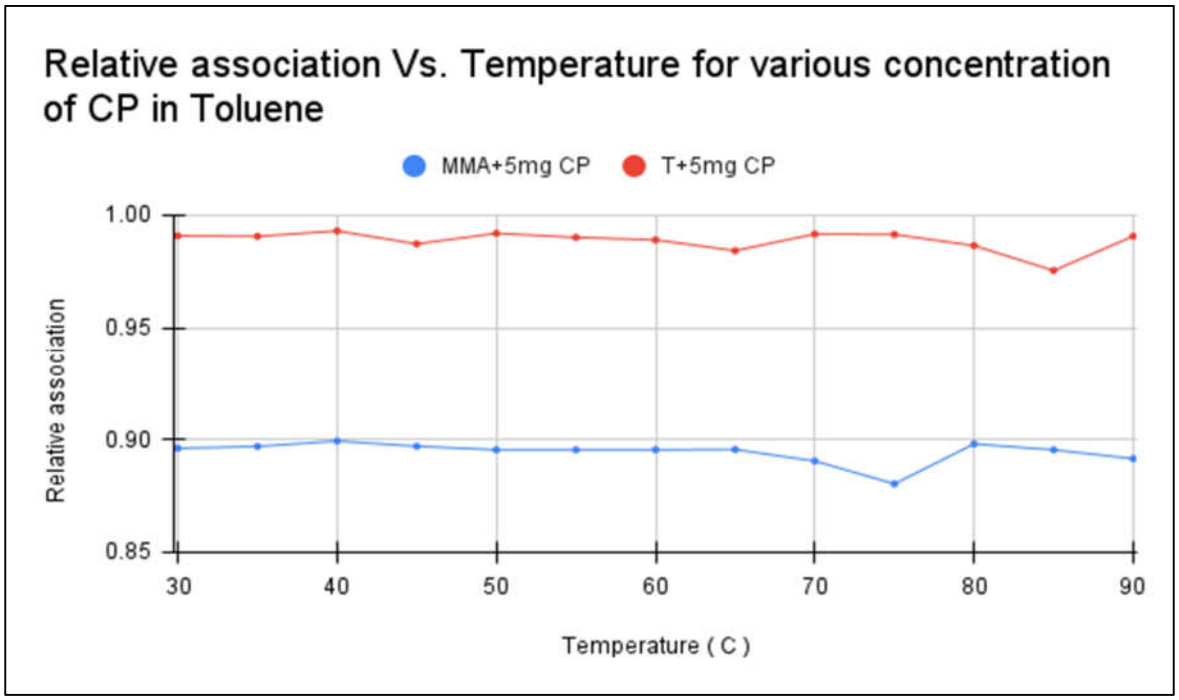

Fig5 Relative association of CLC and PDCLC with temperature

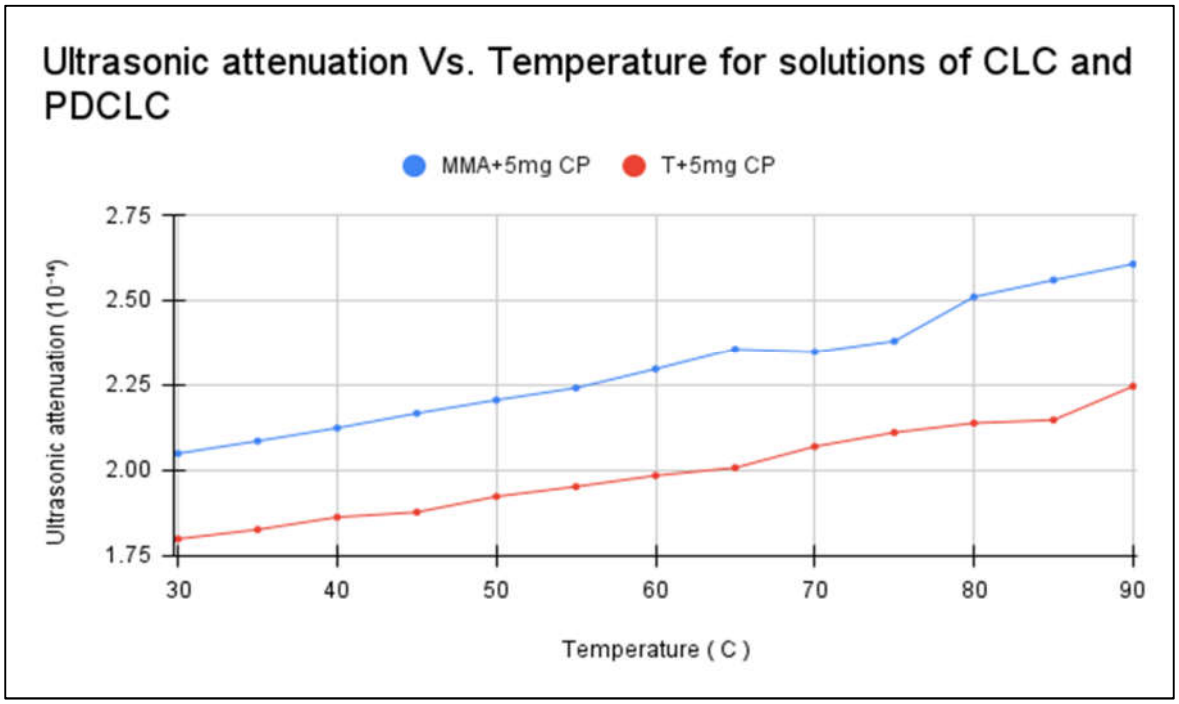

Fig6 Ultrasonic attenuation of CLC and PDCLC with temperature

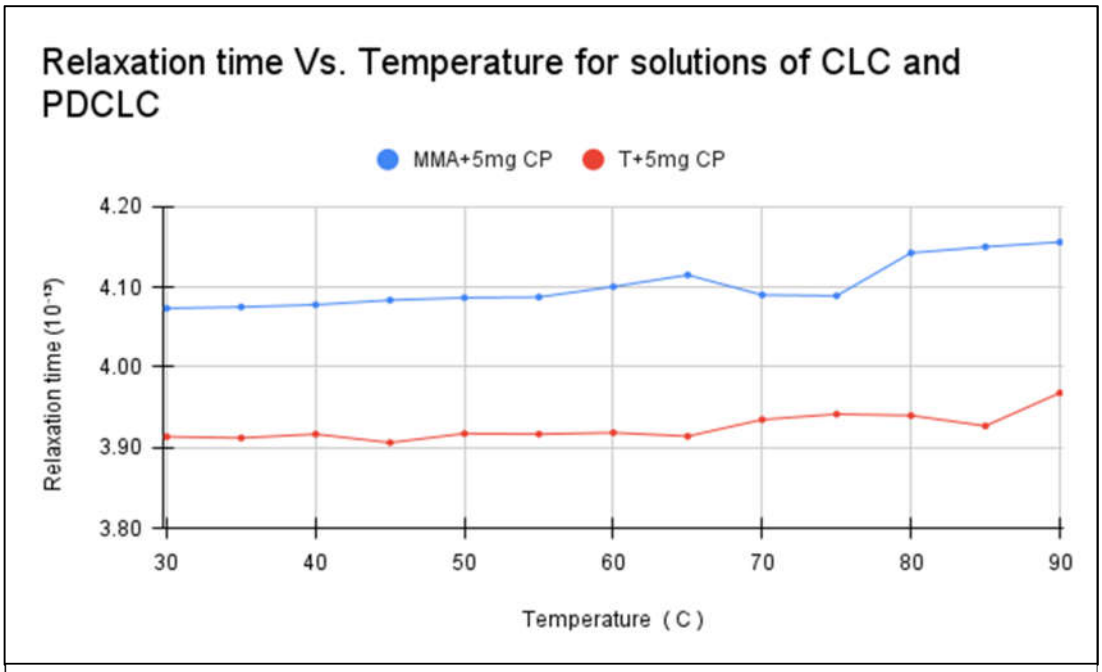

Fig7 Relaxation time of CLC and PDCLC with temperature

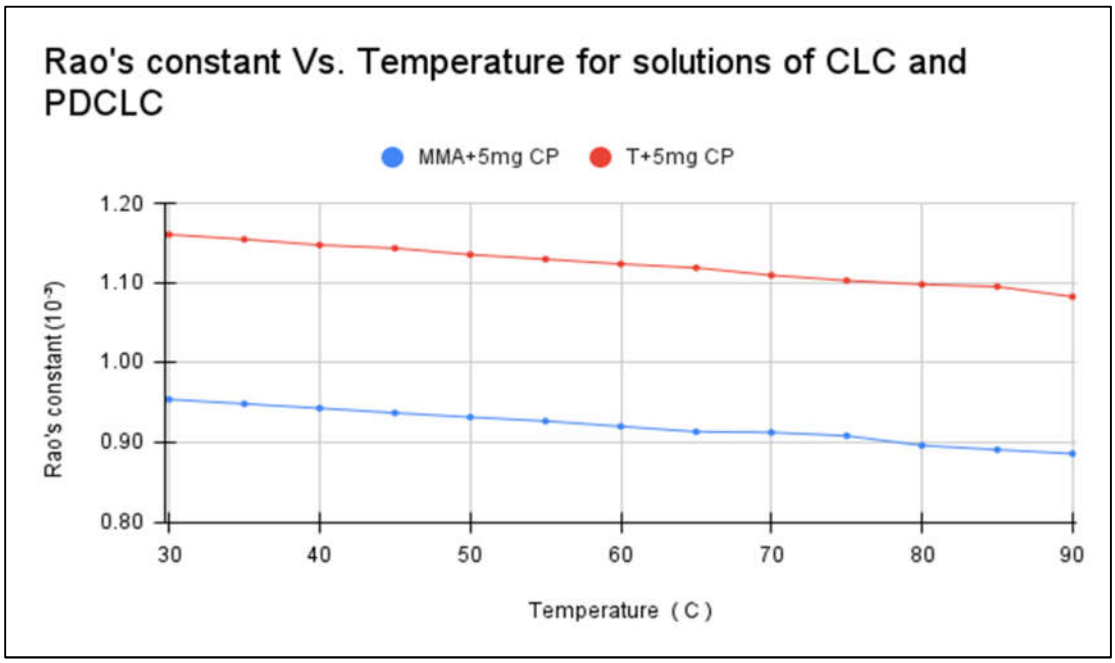

Fig8 Rao's constant of CLC and PDCLC with temperature

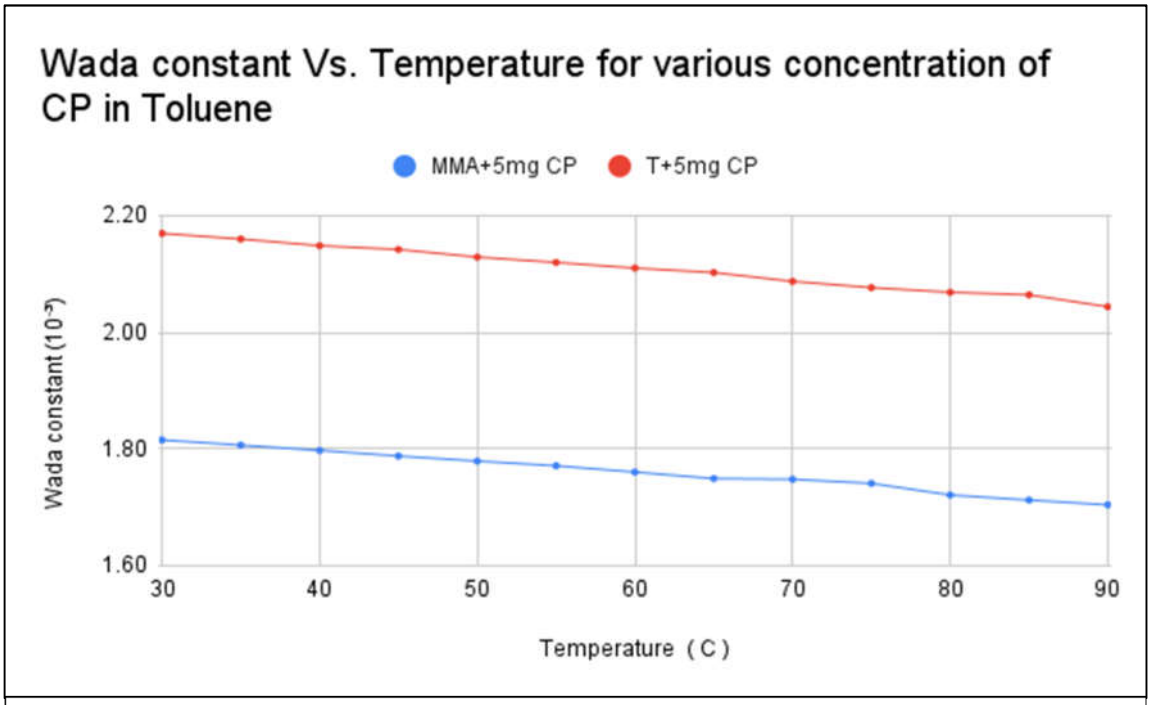

Fig9 Wada constant of CLC and PDCLC with temperature

Discussions:

In both the samples of CLC solution, the sound speed drops as the temperature rises, exhibiting nonlinear behaviour close to the LC mixture PTTs. PDCLC has a lower sound velocity than the combination of toluene and cholesteryl pelargonate. Also, the relative association, Wada constant and Rao's constant decreases in the PDCLC solution. This implies that the polymer's dispersion has enhanced molecular interaction between polymer and LC molecules, resulting in a structural alteration of the composite.

As the temperature rises, the adiabatic compressibility of PDCLC and CP in toluene increases, indicating a decrease in molecular order and an increase in the L_f. PDCLC's adiabatic compressibility is greater than that for CP in toluene, indicating interaction between LC molecules and polymer networks. The ultrasonic attenuation, relaxation time, acoustic impedance also increases in the solution of PDCLC.

The dispersion of LC in the polymer matrix has changed the acoustical behaviour of the pure LC sample, as demonstrated by the graphical fluctuation of acoustical parameters as a function of temperature for both pure LC and PDLC solutions. Comparing PDLC to pure liquid crystal samples, at room temperature, the percentage increases in adiabatic compressibility, L_f, ultrasonic attenuation, and time relaxation are 7.82, 3.99, 12.22, and 3.94, respectively. Comparing PDLC to pure liquid crystal samples, at room temperature, the percentage decreases in velocity of sound, relative association, Rao's constant, and Wada constant are 9.43, 10.56, 21.75, and 19.53, respectively.

Table 3 lists the % change in sound velocity and associated thermodynamic and acoustical parameters of PDCLC.

Table 3 % variation in acoustical parameters of PDCLC compared with pure CLC

Temp (C)	% decrease velocity	% increase β	% increase L_f	% variation Z	% decrease RA	% increase $\frac{\alpha}{f^2}$	% increase τ	% decrease R	% decrease W
30	9.43	7.82	3.99	0.86	10.56	12.22	3.94	21.75	19.53
35	9.61	8.12	4.14	0.70	10.43	12.43	4.01	21.82	19.59

40	9.49	7.93	4.05	0.80	10.41	12.29	3.96	21.78	19.55
45	10.39	9.42	4.83	-0.01	10.05	13.36	4.35	22.11	19.83
50	9.92	8.65	4.42	0.41	10.77	12.80	4.15	21.94	19.69
55	10.00	8.77	4.49	0.34	10.57	12.89	4.18	21.96	19.71
60	10.60	9.75	5.00	-0.20	10.44	13.60	4.44	22.18	19.89
65	11.64	11.42	5.89	-1.14	9.89	14.80	4.89	22.56	20.21
70	9.14	7.33	3.73	1.12	11.35	11.86	3.81	21.65	19.44
75	8.71	6.59	3.35	1.51	12.61	11.34	3.62	21.48	19.31
80	11.65	11.45	5.90	-1.15	9.84	14.82	4.90	22.57	20.22
85	12.80	13.24	6.85	-2.19	8.93	16.11	5.38	22.99	20.57
90	10.79	10.07	5.17	-0.37	11.11	13.83	4.53	22.26	19.95

Results:

The elastic properties of polymer-dispersed liquid crystals as a function of temperature are investigated using an ultrasonic interferometer. The configuration of the CLC material has changed due to the polymer dispersed into it using the polymerization-induced phase separation approach. The incorporation of polymer into the cholesteric liquid crystal reduces the clearing temperature from 91°C to 70°C.The reduction in the clearing temperature of the liquid crystal mixture will enable the liquid crystal solution to operate efficiently at temperatures around room temperature. Optoelectronic equipment, including sensors, filters, and smart windows, will benefit from a low clearing temperature, since less energy will be necessary to transition from an anisotropic to an isotropic state. Consequently, the incorporation of MMA into CLC facilitates precise modulation of these transition temperatures, while preserving the characteristics of pure CLC. The right polymer combination in the CLC could produce a new composite with technological and industrial uses. To comprehend and use cholesteric liquid crystals in a variety of optical and photonic applications, one must be aware of their thermodynamic characteristics, which include phase transitions, acoustic velocity, intermolecular free path length and adiabatic compressibility.

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