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Interdependency among electro-optic characteristics and absorption coefficient of homeotropically aligned liquid crystal considering Beer's law theory

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Abstract. In the present study, interdependency among electro-optic (E-O) characteristics and absorption coefficient of homeotropically aligned liquid crystal (HALC) considering Beer's law theory has been studied. Specifically, HALC cells were prepared using uniform dispersion of ZnO nanoparticles into the nematic liquid crystal without applying any surface treatment to the substrates. Then azo dichroic dye (orange) was added in a fixed concentration to the prepared nanoparticles dispersed LC mixture. The morphological and absorption characteristics of prepared HALC cells were found to be improved with the addition of azo dichroic dye. The optical textures observed at macroscopic and microscopic levels are in correspondence with each other and showed excellent OFF/ON switching with magnificently dark and bright views. Moreover, HALC cells doped with azo dichroic dye exhibited a higher value of absorption coefficient with an improved contrast ratio. Thus, a good consistency was observed among experimentally observed E-O characteristics and theoretically measured absorption coefficient.

Keywords. Liquid crystal; nanoparticles; homeotropically aligned liquid crystal; azo dichroic dye.

1. Introduction

Liquid crystals (LCs) are an intermediate state among solids and liquids phase via sharing properties of both phases [1–3]. Till date, varieties of LC display devices of a high degree of resolution, low energy utilization and slimness have been prepared and used [4–8]. These display devices generally possess homogeneous and homeotropic alignment (HA) of LC molecules. In the history of LC displays, researchers have reported several methods for the alignment of LC molecules on the substrate surface and prepared twisted nematic [9,10], in-plane switching [11], homeotropically aligned [12–14] and fringe field switching [15–17] displays. Among all these, displays based on HA proved to be the most recommended because HA of LC molecules in assembled cells showed signs of much-improved electro-optical (E-O) responses in view of superior contrast, wide view angle, quick response time, outstanding image quality and unessential mechanical rubbing procedure for the molecular alignment [17,18]. In recent times, various groups made use of a variety of nanoparticles (NPs) such as fullerenes [19], gold NPs [20], Ni NPs [21] and POSS NPs [22–24] for inducing HA in LC molecules spontaneously, as this technique was very simple and easy to use via direct doping of NPs into LC mixture [21,23,25–30]. More recently, polyimide-free HA of LC molecules is also achieved using a binary mixture of nematic LC and reactive mesogen [31], silver nanowire networks and nonionic amphiphiles [32], alkylated graphene oxide [33] and nanopatterned organic/inorganic hybrid thin films prepared with polyimide and tin oxide [34].

Moreover, some recent research was also found with guest–host coloured displays with better display properties such as much wider view angle, outstanding shades and high brilliance levels. For getting brilliant and pure colours in guest–host displays [35], the dyes like tetrazines [36], naphthalenes [37], perylenes [38], azo, anthraquinone and acenequinones [39,40] with appropriate characteristics were used. Among these varieties of dyes, azo dichroic dyes have an elongated and rod-like molecular shape that makes it suitable for aligning themselves along with rod-like LC molecules and are preferred for display devices [41]. Consequently, in the present work, the combination of sphere-shaped ZnO NPs with azo dichroic dye is used for the fabrication of homeotropically aligned liquid crystal

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(HALC) and dye-doped HALC cells. The impact of azo dichroic dye on absorption and E-O properties of assembled cells is studied and reported, as azo dichroic dye doped with LC system is responsible for significant absorption in visible range of light in addition to LC molecules. However, the strong absorption in transmission direction can be achieved via appropriate alignment of azo dichroic dye molecules with LC molecules. Thus, parallel alignment of major axis of the molecule (azo dichroic dye/LC) with an electric vector of incident light leads to strong absorption and vice versa. Overall, considering all these factors, absorption coefficients have been measured by taking Beer's law theory under consideration. Efforts have also been made to establish an agreement between experimental and theoretical results.

2. Theoretical phenomenon of absorption study

The transmission properties of HALC cells are strongly affected by the absorption of light via azo dichroic dye molecules. As per the theory of absorption, the strong absorption of light is the result of parallel alignment of the longer axis of molecules with the direction of polarization of incident light, whereas perpendicular alignment leads to weaker absorption. Thus in the present study according to Beer's law, the theoretical expression for OFF-state transmittance (T_{OFF}) of HALC and azo dichroic dye-doped HALC can be found with the parameter extinction coefficient (γ) [42–45] i.e.,

$$\gamma = \alpha + \beta, \tag{1}$$

The 'α and β' correspond to the 'scattering and absorption coefficient' of samples, respectively. Here, T_{OFF} will be expressed as:

$$T = \frac{I_t}{I_0} = e^{-(\alpha+\beta)d}. \tag{2}$$

The terms I_t and I_0 are corresponding intensities of transmitted and incident light. The thickness of the sample is represented by 'd'. As LC molecules and NPs show negligible absorption of light, only azo dichroic dye molecules are responsible for the strong absorption of light in HALC cells. Moreover, the expression for β can be expressed by

$$\beta = \varepsilon x l. \tag{3}$$

Here, symbols 'ε' and 'x' represent 'extinction coefficient' and 'concentration of dopant', respectively. The factor l shows the distance covered by light divided by d and thus $l \approx 1$ corresponds to negligible scattering. Hence according to Beer's law, T_{OFF} for HALC cells is given by:

$$T = \frac{I_t}{I_0} = e^{-(\gamma d)}. \tag{4}$$

3. Materials and methods

A nematic LC mixture MJ98468 (Merck, UK) with negative dielectric anisotropy ($\Delta\varepsilon = -4$) having $n_o = 1.4742$ and $n_e = 1.5512$ as ordinary and extraordinary refractive indices, respectively, have been used as a host material for the preparation of the NPs/dye-doped mixture. The isotropic temperature of pure nematic LC mixture is 75°C. In the given study, spherical ZnO NPs with the average size of particles 50 nm were purchased from SIGMA-ALDRICH and used as collected from the company as one of the dopant materials. Further, another doping material, the azo dichroic dye was also purchased from SIGMA-ALDRICH and used as-received. Azo dichroic dye consisting of (–N=N–) linkage group is also being used commercially for the production of coloured compounds. Moreover, it is highly compatible in LC hosts due to better optical and alignment properties resulting in exhibiting lively (bright) colours with better contrasts by LC displays. Initially, the LC-NPs mixture for HALC cell was prepared by well dispersing the specific amount of sphere-shaped ZnO NPs (0.3 wt/wt% to LC) at the isotropic temperature by overnight stirring using a hot plate and stirrer. This is worth to mention that the experiments were performed for different concentrations of NPs. Considering the experimental observations such as homogeneous dispersion without any visible aggregation and repeated as well as reproducible E-O results, the specific concentration was found the most optimized value. For supportive evidence, the authors have also discussed the detailed mechanism of achieving homeotropic anchoring in earlier published reports in the case of pure LC system by taking the same optimized concentration of NPs [3,14]. Then, azo dichroic dye-doped sample was prepared by homogeneous mixing of 0.0625% (wt/wt) of azo dichroic dye in the prepared LC-NPs mixture by continuously shaking and heating simultaneously to its isotropic temperature. Thereafter, glass substrates coated with indium tin oxide (ITO) were used for the fabrication of two different cells each of having a thickness (cell gap) of 10 μm. It is to be noticed that the percentages of LC material, NPs and azo dichroic dye used for HALC and dye-doped HALC cells were [99.70% + 0.3% + 0%] and [99.63% + 0.3% + 0.0625%], respectively. In order to examine the aligning behaviour of LC molecules over ITO-coated substrate surface, the morphology of the surface was studied by taking optical textures by means of a polarized optical microscope (POM; Nikon LV100POL, Japan). Further, the E-O properties of prepared cells positioned among crossed polarizers, at a direction of 45°, were studied by making use of a laser beam with wavelength 632 nm and a square wave at a frequency of 60 Hz using a function generator (AFG 3021B, Tektronix, USA). Additionally, to study the effect of the ZnO NPs and azo dichroic dye on the phase transition temperature of LC, the HALC cells were studied using a programmable temperature controller (Model TP 94 and THMS600E, Linkam, UK) at a constant rate of 0.5°C min^{-1}.

4. Results and discussion

4.1 *Macroscopic view of HALC cells in ON and OFF states*

Textural study of HALC and dye-doped HALC cells prepared via doping of ZnO NPs and azo dichroic dye was carried out using POM with respect to the applied voltage. In addition, the macro textures of the prepared cells were investigated and recorded with the help of high-definition digital camera, as shown in figure 1, where images (a–d) and (e–h) reflect the textures for HALC cell and dye-doped HALC cells, respectively. Especially, the active area is highlighted in all images (a–h). Textures in figure 1 (a and e) in OFF state (at 0 V) represent the black state for both samples, which gives a clear indication about the homeotropic alignment of LC molecules. Further, on applying voltage in ON state, both cells in figure 1b–d and f–h show changes in state from black towards white, which reflect the orientation of LC molecules with the applied voltage. Moreover, the basic behaviour about a change of state for cells (under investigation) on applying voltage remains the same but the switching voltage varies for both cells. A lesser voltage is required for dye-doped HALC cell to start switching from the black state as seen clearly from textures of figure 1b and f (1.65 < 1.70 V). Moving further, textures of figure 1c and g again show similar results; i.e., 1.75 < 1.8 V. Similarly, on applying more voltage, the orientation of LC molecules get saturate and the textural images become maximum white, as shown in figure 1d and h, where azo dichroic dye-doped sample shows much better macroscopic bulky state at a comparatively lower voltage (1.9 < 2.0 V).

4.2 *Microscopic textural behaviour of HALC cells in ON and OFF states*

In addition to the macroscopic textures study of prepared cells, the optical textures were also captured at a microscopic level with the help of POM, where crossed polarizers were set at a magnification of 10X (shown in figure 2). The POM textures in OFF state (figure 2a and e) show a perfect dark state due to the perfect HA of LC molecules with respect to the substrate. However, in ON state, the sample doped with an appropriate concentration of azo dichroic dye shows switching from dark to white state at lower voltage, as compared with the HALC sample cell. The requirement of lesser switching voltage for azo dichroic dye-doped sample compared with HALC sample cell may lead to the conclusion that azo dichroic dye molecules as dopant

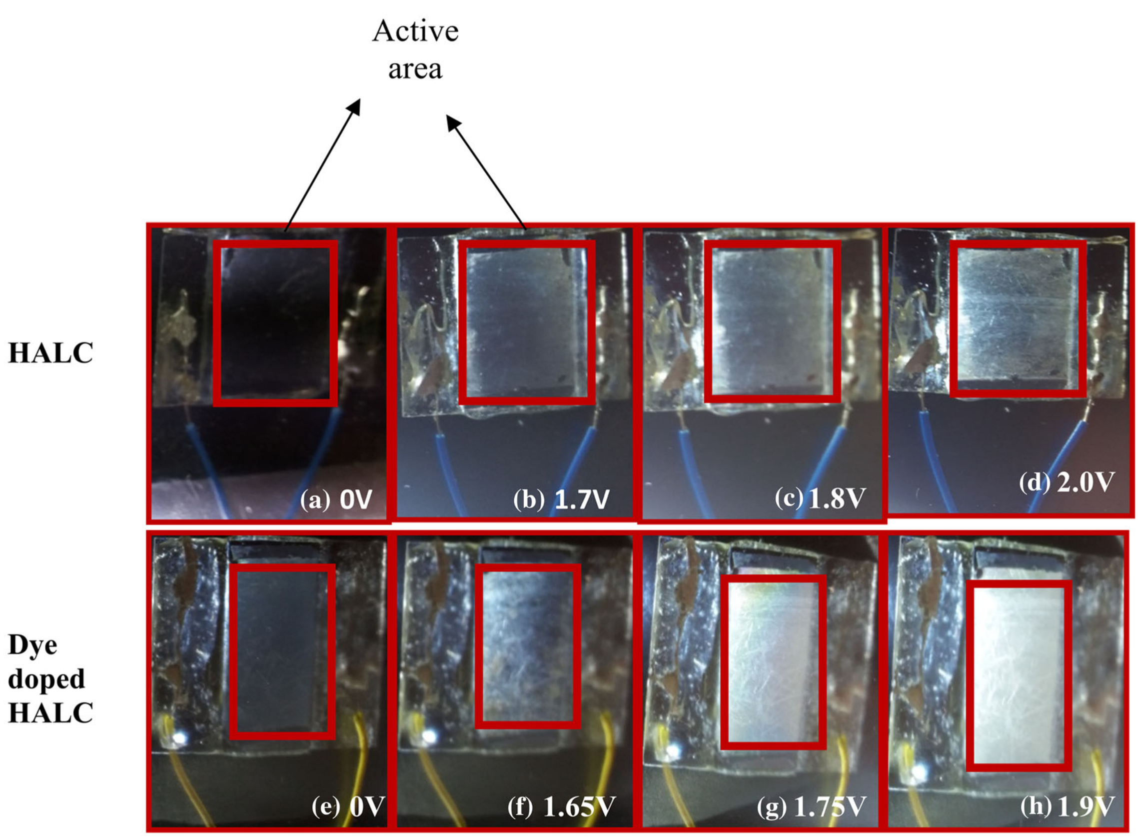

Figure 1. Macroscopic view of (**a**–**d**) HALC and (**e**–**h**) dye-doped HALC cells at different applied voltages.

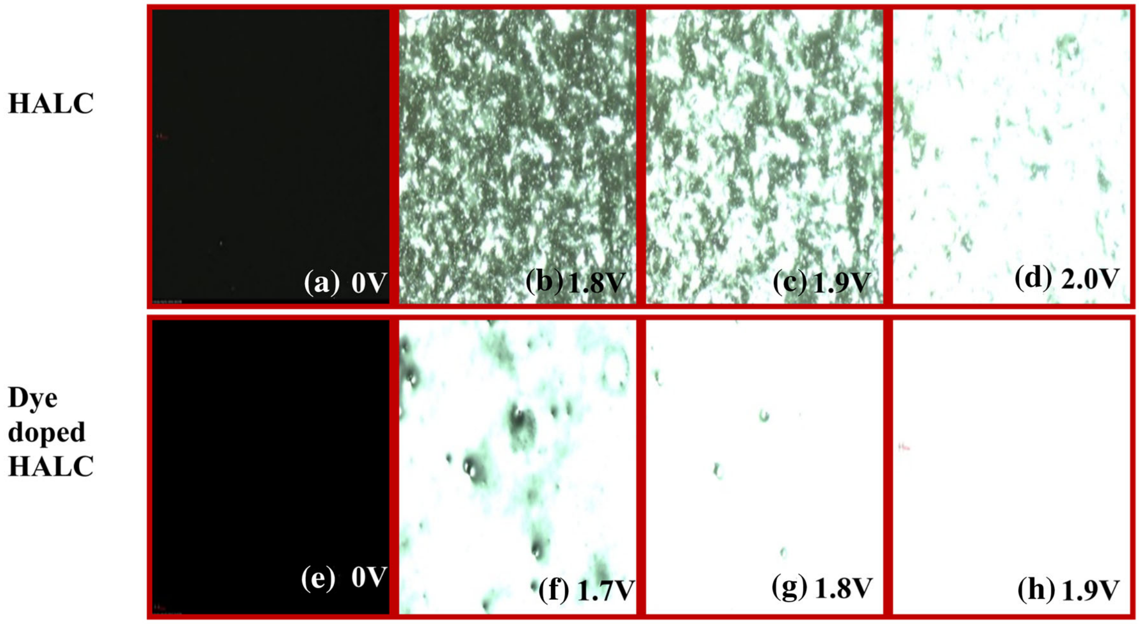

Figure 2. Microscopic textures of (**a–d**) HALC and (**e–h**) dye-doped HALC cells in OFF and ON states.

reduced the anchoring strength due to weakening of LC-dye bond strength [1,2].

4.3 *Absorption coefficient (β) based on theoretical consideration*

Keeping in mind theoretical concern, the dye extinction coefficient (γ) can also be determined by the experimental value of OFF-state transmittance and vice versa by making use of equation (4). For example in the present situation, let extinction coefficient of HALC as γ_1. Further, we assume that there is very little absorption of light by LC molecules in HALC cells because NPs do not absorb light, though this absorption will be negligible. So let here the extinction coefficients of HALC as $\gamma_{1||||}$ and $\gamma_{1\perp}$ in the similar direction corresponding to major and minor axes of LC molecule, respectively. Two cases arise from these coefficients:

Case I. Think about a situation where LC molecules are dispersed in 1D or 3D space, the relation among γ_1, $\gamma_{1||||}$ and $\gamma_{1\perp}$ is given by [42]:

$$\gamma_1 \approx \frac{\gamma_{1||} + 2\gamma_{1\perp}}{3} \approx 0.333\gamma_{1||} + 0.667\gamma_{1\perp} \tag{5a}$$

and

$$\gamma_{1||} > \gamma_1 > \gamma_{1\perp}. \tag{5b}$$

Case II. Now, consider a state for 0.0625% dye-doped HALC cell, the absorption coefficient $\beta_{21||}$ is greater than $\beta_{2\perp}$ due to the strong absorption of light in parallel direction to the major axis of doping material, then

$$\gamma_2 \approx \frac{\gamma_{2||} + 2\gamma_{2\perp}}{3} \approx 0.333\gamma_{2||} + 0.667\gamma_{2\perp} \tag{6a}$$

and

$$\gamma_{2||} > \gamma_2 > \gamma_{2\perp}, \tag{6b}$$

where γ_2 is extinction coefficient of HALC cell doped with azo dichroic dye. Moreover, $\gamma_{2||}$ and $\gamma_{2\perp}$ are extinction coefficients (along major and minor axis) of azo dichroic dye molecule.

Using equations (5b and 6b), we have

$$\gamma_{2||} > \gamma_{1||},\ \gamma_{2\perp} > \gamma_{1\perp} \text{ and } \gamma_2 > \gamma_1, \tag{7}$$

$$\gamma_{2||} > \gamma_{1||} > \gamma_1 \tag{8a}$$

and

$$\gamma_{2||} > \gamma_{2\perp} > \gamma_{1\perp}. \tag{8b}$$

Using inequality (8a) comparable to (5b) in (5a),

$$\gamma_{1||} \approx \frac{\gamma_{2||} + 2\gamma_1}{3}.$$

Now by putting the value of γ_1 from equation (5a) in above equation, we get

$$\gamma_{2||} \approx 2.34\gamma_{1||} - 1.33\gamma_{1\perp}. \tag{9}$$

Similarly, using inequality (8b) comparable to (5b) in equation (5a),

$$\gamma_{2\perp} \approx \frac{\gamma_{2||} + 2\gamma_{1\perp}}{3}. \tag{10}$$

On putting value of $\gamma_{21||}$ from equation (9) in (10), we get

$$\gamma_{2\perp} \approx 0.77\gamma_{1||} + 0.22\gamma_{1\perp}. \tag{11}$$

Using equations (9) and (11) in equation (6a), we have

$$\gamma_2 \approx 1.29\gamma_{1||} - 0.29\gamma_{1\perp}. \quad (12)$$

Normally, T_{OFF} for HALC by using equation (4), is given by:

For HALC

For 0.0625%dye-doped HALC

$$T = \begin{cases} T_1 \approx \exp - (\gamma_1 d) \\ T_1 \approx \exp - (\gamma_1 d) \end{cases} \quad (13)$$

Using equations (5a) and (12) in equation (13), we get

For HALC

For 0.0625%dye - doped HALC

$$T = \begin{cases} T_1 \approx \exp[-(0.333\gamma_{1||} + 0.667\gamma_{1\perp})d] \\ T_2 \approx \exp[-(1.29\gamma_{1||} - 0.29\gamma_{1\perp})d] \end{cases} \quad (14)$$

However, $\gamma_{1\perp} \to 0$, when the direction of polarization is similar to absorbance axis of azo dichroic dye molecule, so T_{OFF} from equation (14) will be expressed as:

For HALC

For 0.0625%dye-doped HALC

$$T = \begin{cases} T_1 \approx \exp[-(0.333\gamma_{1||})d] \\ T_2 \approx \exp[-(1.29\gamma_{1||})d] \end{cases} \quad (15)$$

Thus equation (15) represents the expressions for minimum OFF state transmission (T_1 to T_2) for HALC sample cell doped with 0 and 0.0625% dye concentrations based on theoretical consideration of Beer's law. Equation (15) clearly exhibits that the addition of some concentration of dye reduced the OFF state transmittance, which results in higher contrast ratio (CR).

Further, β can be calculated for various samples by putting values of γ_1 and γ_2 from equation (15) and x as 0.3 and 0.3625 for HALC and 0.0625% dye-doped sample, respectively, in equation (3)

For HALC cell

For 0.0625%dye - doped HALC

$$\beta = \begin{cases} \beta_1 = 0.099\gamma_{1||} \\ \beta_2 = 0.4676\gamma_{1||} \end{cases} \quad (16)$$

Consequently, theoretical computation showed the experimental result of larger value of β with the addition of azo dichroic dye as shown in figure 2, may be due to dye orientation with respect to direction of incident light, which in turn depends upon internal dipole–dipole interaction among the excited dye molecules and nematic LC molecules.

Further, by making use of equations (1), (3) and (4), the value of β for HALC cells was evaluated in OFF state using the experimental value of T_{OFF}. Here, the concentration of doping material was considered as 0.3 and 0.3625, as well as the value of l is approximately unity. Figure 3 represents the absorption coefficient value comparison of both sample cells in OFF state. Results show that the value of β is enhanced with the addition of azo dichroic dye in the sample because azo dichroic dye molecules are much soluble and possess advanced optical as well as alignment properties in host LC.

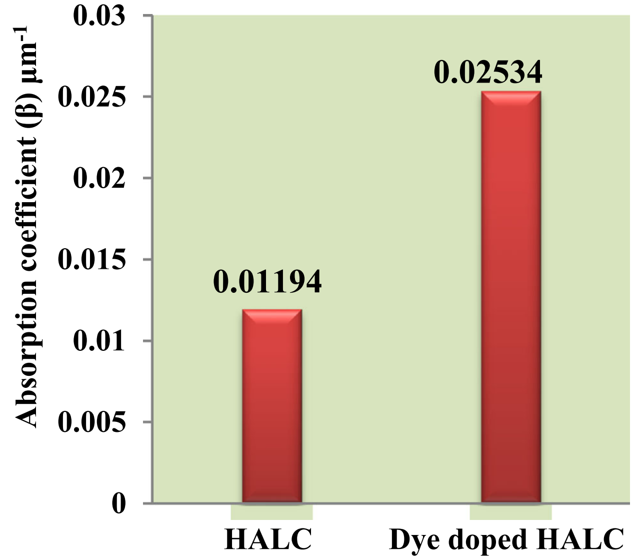

Figure 3. The OFF state values of the absorption coefficient for HALC and 0.0625% azo dichroic dye-doped HALC cells.

4.4 *Transmission behaviour vs. absorption coefficient*

The transmission behaviour of HALC and dye-doped HALC cells were studied with respect to absorption coefficient, shown in figure 4a. The OFF state transmission was also compared for both the cells in figure 4b. Figure 4a shows that the transmission for both cells decreases exponentially with respect to absorption coefficient β that meets with the theoretical expectation from equations (3) and (4). Further, figure 4b represents the comparison of the OFF states' transmissions (T_{OFF}) of cells, according to which T_{OFF} (azo dichroic dye-doped cell) < T_{OFF} (HALC cell) due to increase in value of β simultaneously.

4.5 *Contrast ratio vs. absorption coefficient*

The E-O response of prepared LC cells can also be represented with significant factor CR that depends upon maximum and minimum values of transmittance corresponding to ON and OFF states, respectively. Moreover, theoretically, the expression for CR can be given by equation (17) as:

$$CR = \exp[(\gamma_{OFF} - \gamma_{ON})d] (17)$$

here, 'γ_{OFF} and γ_{ON}' represent 'extinction coefficients in OFF and ON states', respectively, which depends on the

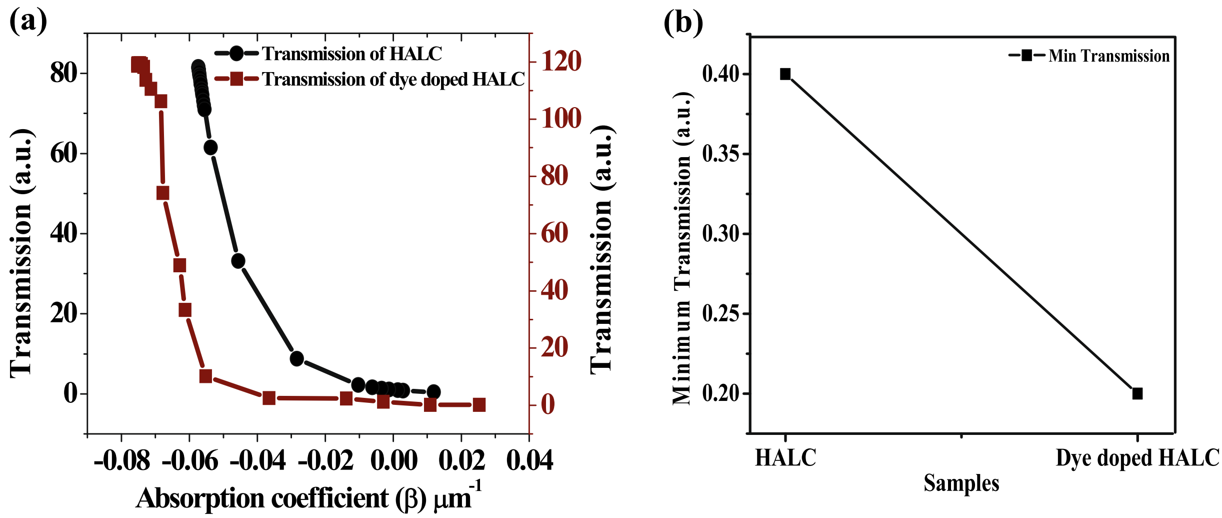

Figure 4. (**a**) Transmission behaviour of HALC cells with respect to the absorption coefficient β. (**b**) The OFF state transmission values of HALC and dye-doped HALC cells.

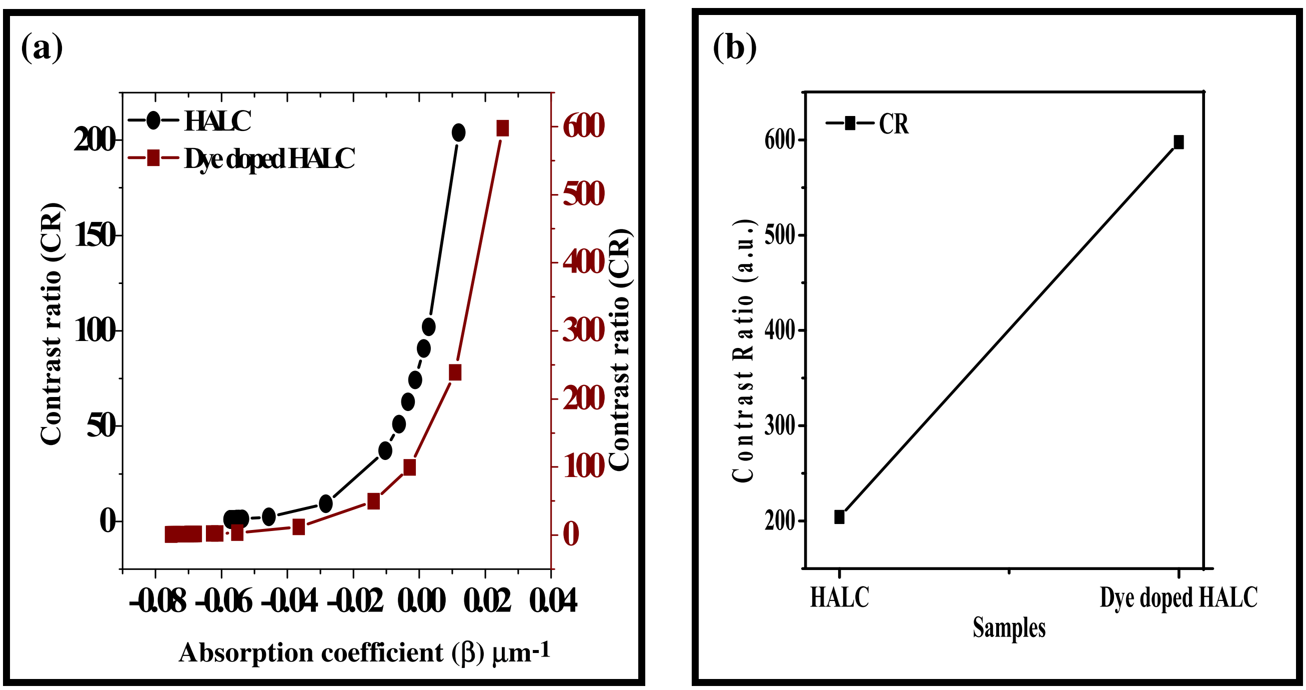

Figure 5. (**a**) Contrast ratio for HALC and dye-doped HALC cells with respect to the absorption coefficient. (**b**) The values of CR for HALC and dye-doped HALC cells.

value of β and d is the cell thickness. Further, complete behaviour of CR has been given in figure 5 with respect to absorption coefficient and shows the exponential improvement in CR with an increase in absorption of light by the molecules for both samples. Moreover, the enhancement in CR is more in azo dichroic dye-doped HALC cells as compared to HALC cells. Further, a comparison of calculated values of CR has been mentioned in figure 5b, according to which azo dichroic dye-doped HALC cell shows much better contrast compared with HALC sample cell, which may be due to lower transmittance value in OFF state as compared to HALC cell. Moreover, from equation (15), it is clear that the azo dichroic dye-doped HALCs have a higher extinction coefficient compared with the extinction coefficient of HALC that corresponds to lowering of T_{OFF} and enhancement in CR.

4.6 *Phase transition behaviour with temperature*

The phase transition behaviour of HALC and azo dichroic dye-doped HALC cells was studied under POM using a hot stage connected with a temperature controller followed by heating and cooling of cells at a uniform rate of 0.5°C min^{-1}. To study the effect of the ZnO NPs and azo dichroic dye on the phase transition temperature of LC with the most accuracy, the HALC and dye-doped HALC cells were heated and cooled several times. The experimentally recorded textures during heating and cooling cycles showed the transition of molecules from nematic to isotropic and vice versa with a significant change in transition temperature for HALC and dye-doped HALC cells. The phase transition temperatures of pure LC, HALC and dye-doped HALC cells were recorded as 75, 83.2 and 76.7°C, respectively. A considerable change in the isotropic temperature under POM was perceived in the HALC cell, which may be due to the formation of NPs layers deposited on ITO substrates in the confined cell that make it more stable to the cell with respect to temperature. Whereas it seems that the absorption of dye molecules leads to the reduction in phase transition temperature in dye-doped HALC cells.

5. Theoretical and experimental consistency in results

Based upon Beer's law, the absorption coefficient was calculated theoretically in equation (16) and accordingly the value of β was found to be increased with the doping of azo dichroic dye into the host sample. Further, experimentally, on the basis of transmission study, the values of β were determined and are reflected in figure 3, which also show consistency with the theoretical observation of β *vs.* azo dichroic dye. Moreover, theoretically, equation (4) shows the inverse relation between transmission and absorption, as analysed experimentally and also given in figure 4a. Additionally, the CR was studied using equation (17), which showed the enhancement in CR with the addition of azo dichroic dye and this theoretical improvement, in contrast, lies in good agreement with the experimentally observed behaviour of CR, as shown in figure 5a and b. Thus, theoretical calculations for determining the behaviour of absorption coefficients are in consistency with the experimental results found with the studies of the same.

6. Conclusion

A theoretical model based on Beer's law was taken under consideration to study the impact of azo dichroic dye on E-O cum absorption characteristics of prepared sample cells. Two sample cells were prepared using 0 and 0.0625% azo dichroic dye in host LC as well as studied. The theoretical calculation was made using Beer' law equations for the determination of transmittance, absorbance and CR. The optical textures observed at macro and microscopic levels show the good dark and bright states under off and on voltages, respectively. Further, good consistency was observed among experimentally observed E-O characteristics such as minimum and maximum transmission, CR as well theoretically measured absorption coefficient. In addition, the temperature-dependent phase transition behaviour of HALC cells showed a significant effect of the NPs and azo dichroic dye on the isotropic temperature. The phase transition temperatures of pure LC, HALC and dye-doped HALC cells were observed as 75, 83.2 and 76.7°C, respectively, in POM study.

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