U.P.B. Sci. Bull., Series B, Vol. 87, Iss. 4, 2025 ISSN 1454-2331

INVESTIGATION OF BQMI ZWITTERIONS-LIQUID CRYSTALS COMPOSITES IN VARIOUS POLYMER MATRICES FOR PDLC APPLICATIONS

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Polymer-Dispersed Liquid Crystals (PDLCs) are a versatile class of materials widely investigated for applications in optical devices such as smart windows. The response of these materials relies on the electrically induced realignment of liquid crystal domains dispersed within a polymer matrix, process that is reversible upon the removal of the electric field.

In this study the benzoquinonemonoimine (BQMI) zwitterion, known for its large internal dipole moment and favorable charge transport properties, was incorporated alongside the nematic CB5 liquid crystal in four distinct polymer matrices. The key role of the zwitterions as a dopant is to enhance the electro-optical response of the final materials.

Four formulations were developed using CB5, BQMI, and polycarbazole-based matrices, and the resulting PDLCs were evaluated to determine the effects of composition on morphology and switching behavior. The findings provide insight into the role of zwitterionic doping as well as the interaction between the polymer structure and liquid crystal in tuning the performance of PDLC systems.

Keywords: zwitterions, polycarbazole, polymer-dispersed liquid crystals

1. Introduction

Liquid crystals (LC) are materials characterized by their structural flexibility and a high degree of order across a certain macroscopic direction which

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is called director. This property comes as a result of the intermolecular interactions, van der Waals interactions and the strong intrinsic polar character. [1-3]

The abovementioned traits of LC led to their use in various types of devices such as smart windows, sensors and artificial muscles.[4,5] Most state-of-the-art applications or medical technologies, liquid crystal mixtures are used to generate favorable ”group” properties by mitigating individual specific limitations. New technological applications require the design of new LC structures characterized by specific properties. Due to the fact that chemical synthesis is expensive from a technological and temporally point of view, a more elegant way is to manufacture LC-based mixtures. To achieve this, additions of new components are used to regulate and guide macroscopic properties for the desired application. [6-10]

It was demonstrated that the use of thermo/solvato/photo/ (pre)polymer additions creates a new stable LC system. Moreover, using a polymer into the LC matrix does not interfere only at management level of electro-optical properties, but also offers robust mechanical support for LC molecules (structural stabilization), giving flexibility to the obtained composite films. [9-13]

Based on light transmission control (at a particular applied voltage value) polymer disperse liquid crystals (PDLC) represent a class of materials with wide utility in the design and manufacturing of smart windows.[14-17] Recent efforts were done in order to solve the disadvantages of PDLCs, particularly the low control voltage. [18-21]

Carbazole has proven to be a versatile platform for developing materials with applications in various domains ranging from optics to pharmaceuticals. Due to their stability and aromatic character, polycarbazole based compounds have high thermal and photochemical resistance. [22]

The electrical properties of carbazole materials, influenced by their conduction and valence band potentials, can be further tuned through the insertion of different substituents on the aromatic ring of the carbazole. [23-26]

These specific features make the carbazole molecules important candidates for the development of new energy storage materials such as solar cells and for the synthesis of a wide range of 1) dyes with strong absorption profile [27,28], 2) materials with hole conduction [28,29], molecular glasses [30-32] and 3) light emitters. [33-36]

Based on this information and the previous studies carried out in the laboratory of the synthesis of polymers with special properties, from Faculty of Chemical Engineering and Biotechnologies, the main purpose of this paper is to manufacture PDLC assemblies based on LC and carbazole derivatives, using as dopant stabilizer-energy stoker small organic molecules with zwiterionic nature.

2. Materials and Methods

The polymer matrices used in this study are: a) poly(N-acryloylmorpholine, Mn= 29 KDa) **I**; b) poly(N-acryloyl morpholine-co-MK); **II:** R = R_1= CN; **III**: R = H; R_1= NO_2; c) poly (N-acryloyl morpholine-co-N-vinyl, N-acetamide co-MK) **IV** (Fig. 1. a-c). These polymers were synthesized in our laboratory. [37-39]

The benzoquinonemonoimine zwitterion (BQMI, Fig. 1. d) was kindly provided by the Centre Interdisciplinaire de Nanoscience de Marseille [40] (CINaM-UMR CNRS 7325), Aix-Marseille Université and the liquid crystal 4-pentyl-4'-cyanobiphenyl (5CB) was purchased from Merck (Fig. 1. e). All the other reagents were acquired from commercial suppliers and used as received.

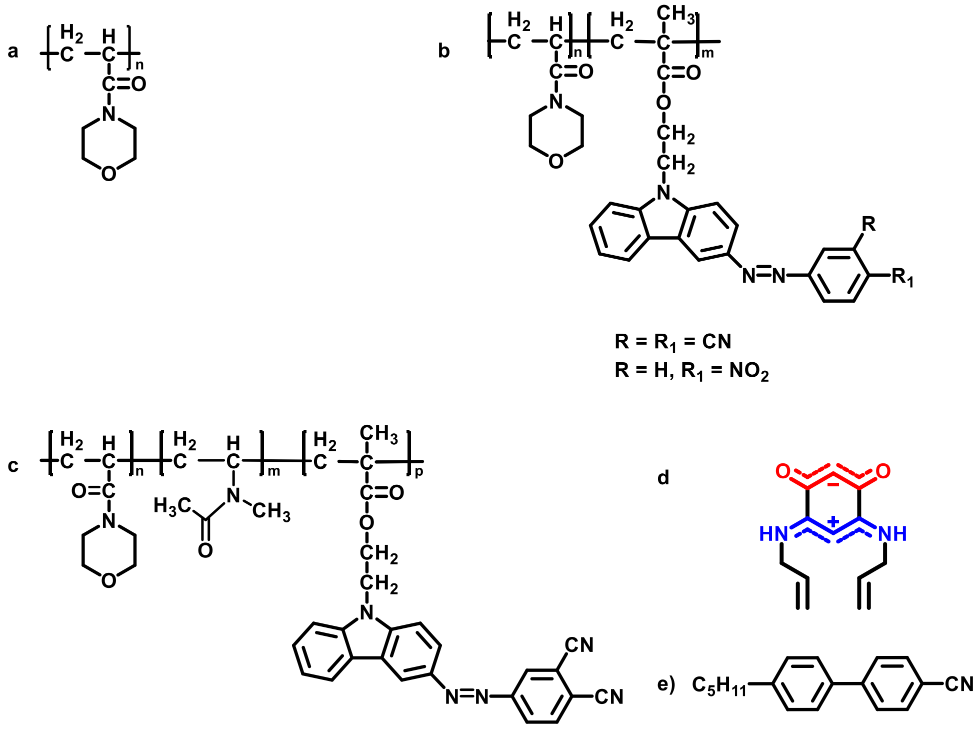

Fig. 1. Chemical structures of PDLC components: a) **I**: polyNAM; b) **II**: R = R_1= CN; **III**: R = H; R_1= NO_2; c) **IV** d) zwitterion; e) 5CB

For PDLC preparation, the zwitterion (15% reported to the LC) and 5CB (34% reported to the polymer) were dispersed in a $CHCl_3$ solution containing the polymer matrix (10%).

The cells employed for electro-optical measurements (Fig. 2.a) were filled by capillarity with PDLC composition at the specific temperature for isotropic phase of 5CB (higher than 35.1°C).

The setup employed for the electro-optical characterization of PDLC cells (Fig. 2.b) consisted of a Tektronix AFG3022C arbitrary function generator, which provided a burst sinusoidal waveform with an amplitude of 1-20V and a frequency of 1 kHz, further amplified 20× using a Tegam 2350S high voltage amplifier. A laser beam (λ = 633 nm) emitted by a ThorLabs HNL020L He-Ne laser was transmitted through the cell towards a ThorLabs PDA36A-EC silicon amplified photodetector, which provides an output voltage proportional to the incident light intensity. The photodetector output voltage was recorded using an Agilent DSO7014B digital storage oscilloscope.

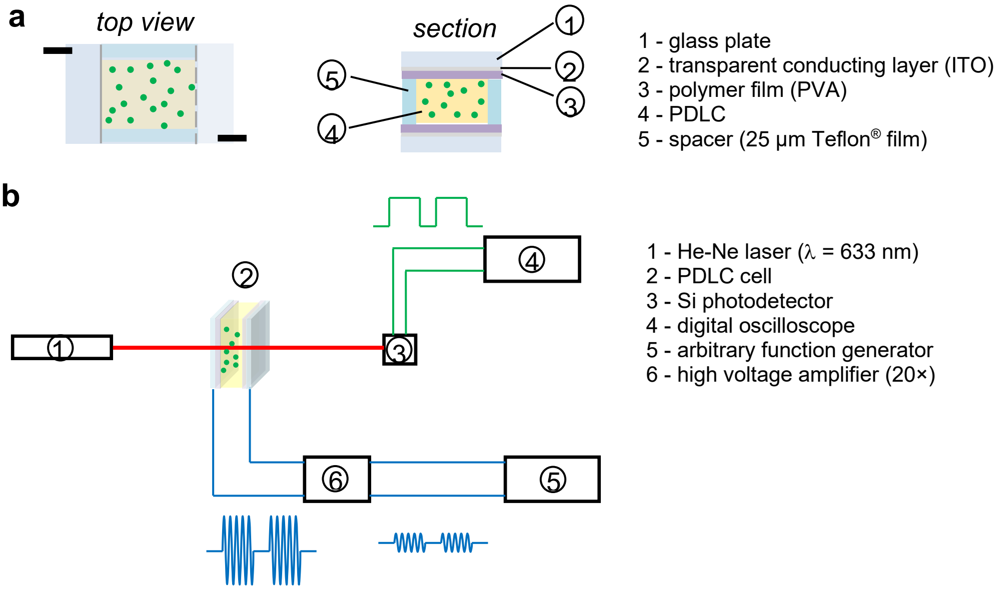

Fig. 2. a) Representation of a PDLC cell; b) Electro-optical characterization setup.

Absorption spectra of PDLC films on glass slides, in the UV-visible region, were recorded using a Jasco V550 spectrophotometer at room temperature.

3. Results and discussion

3.1. UV-VIS characterization

The UV–Vis spectroscopy is a characterization method which offers the first information for molecular transitions. Moreover, the specific structural interactions between the different components of composites could be highlighted by the comparative analysis of all materials answers. [33]

Thus, Fig.3. presents the recorded spectra for the four compositionally derived compositions (Table 1.), for a better understanding of the interactions between components.

Table 1:

Composition of PDLC materials

Compositions	Polymer matrix	Dye	Liquid Crystal
A	poly(N-acryloyl morpholine-co-MK) **II**	Zwitterion	CB5
B	poly(N-acryloyl morpholine-co-MK) **III**	Zwitterion	CB5
C	poly (N-acryloyl morpholine-co-N-vinyl, N-acetamide co-MK) **IV**	Zwitterion	CB5
D	poly(N-acryloylmorpholine) **I**	Zwitterion	CB5

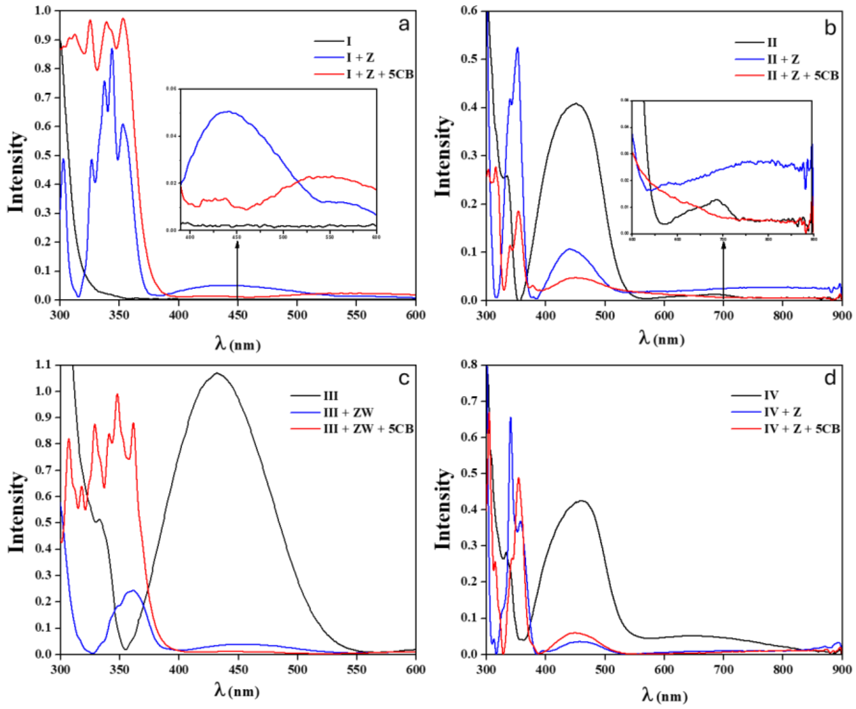

Fig. 3. Electronic absorption spectra of analyzed precursor and PDLC compositions a) pNAM **I**; b) p(NAM co I) **II**; c) p(NAM co II) **III**; d) p(NAM co NMNVA co I) **IV**; black line: only polymeric matrix; blue line: polymeric matrix doped with zwitterion; red line: PDLC composition matrix-zwitterion - 5CB (A-D)

In all analyzed cases the electronic spectra of the polymer films present particular absorption maxima centered around: 340 nm for the composition containing **I** as a polymer matrix (Fig.3. a)) and 450 nm for the other three compositions (Fig. 3. b-d)). The change in their position and intensity are closely related to the composition of the material. Thus, for the basic constituent of all materials, pNAM, as it was already demonstrated in previous studies, no absorption is recorded in the visible region. This behavior is the consequence of its structure with low molecular dipole value, due exclusively to the difference in electrochemical character, induced by the presence of N and O heteroatom's, not involved in conjugation/resonance structures (Fig. 3. a). [29] The presence of the zwitterion (blue curve, Fig. 3 a)) generates absorptions in the n-π* transition region, respectively 442 nm specific to the zwitterion. [41] By adding LC the maximum absorption gains in intensity - hyperchromic effect in the region 350-400 nm; simultaneously with the hypochromic red-shifted: λ_{max} centered around 550 nm (bathochromic effect) (red curve, Fig. 3a)). This change demonstrates charge quenching by intermolecular transfer between zwitterion and LC (charge complex transfer-CTC).

Significant changes were observed after 300 nm for the copolymers (Fig 3. b-d). The electronic band located at 400-500 nm is due to a strong interaction between randomly distributed chromophores (azocarbazole moieties), zwitterion and LC, respectively. [34,35,39,42]

This response is attributed to the trans-cis photoisomerization of the azoaromatic residues. The electronic transition of the azoaromatic sequence is influenced by the interaction between the zwitterion and the liquid crystal complex.

Thus, the absorbance of PDLC composites is a consequence of the cooperative dipole–dipole interaction between the structural polar moieties of the components. In addition, a peculiarity of the azo-dicyan derivative (black curve, Fig. 3b), although weak, is the absorption peak in the entire visible range. It appears that the resulting dipole at the level of the two cyanide sequences in the polymer structure (Figure 1. c) favors the panchromatic absorption, mainly due to the change in the electronic structure of the zwitterion in the presence of LC.

3.2. Polarized light microscopy

Under microscopic examination in polarized light, nematic liquid crystals display typical schlieren wire and marbled textures while cholesteric, smectic or discotic LCs have conic focal, polygonal, fan and mosaic textures. Homogeneous nematic films generated in cells with treated surfaces are involved in temperature- and field-induced orientational effects, including Fredericks or local Fredericks transition.

Stabilization of nematic phases can be achieved by polymer matrices that generate liquid crystal domains with relatively uniform dimensional distribution. Such interconnected cross-linking domains can be generated in preformed polymer matrices by evaporation of a common solvent or by promoting polymerization in the monomer-liquid crystal mixture.

The generated polymer-liquid crystal assembly is characterized by its sensitivity towards thermal, electrical or electromagnetic stimuli, which is a consequence of the LC interaction with the other components of the material, stabilized in the matrix. In this way, the influence of the molecular structure of the polymer affecting the molecular orientation of the LC is observed. If the polymer structure resembles to that of the LC molecule, the molecular orientation of the LC molecules around the polymer is maintained as shown in Fig. 4. a), and the LC exhibits a nematic state. In contrast, the molecular orientation of the LC molecules around the polymer is disrupted if the molecular structure of the polymer is not close to that of the LC molecule as illustrated in Fig. 4. b). In this case the nematic to isotropic transition temperature (T_{NI}) is finally shifted.

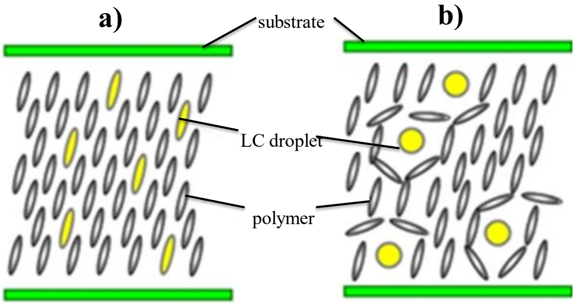

Fig. 4. Schematic representation of the orientation for component molecules in PDLC compositions: a) matrix with similar molecular structure to that of LC b) matrices with a different structure from LC.

Hot stage polarized light microscopy (POM) is a characterization technique which allows simple initial determination of liquid crystal droplet segregation and, in addition, provides information about the phase behavior and the corresponding transition temperatures.

A preliminary POM investigation of PDLC cells was carried out at room temperature under crossed polarizers and revealed that PDLC composites consist of bright-colored anisotropic domains containing LC distributed uniformly in the isotropic polymer matrix (Fig. 5.). The difference in size and number of domains is due to the miscibility of 5-CB with the various polymer matrices, and the tendency to segregate decreases in order D < B ~ C< A.

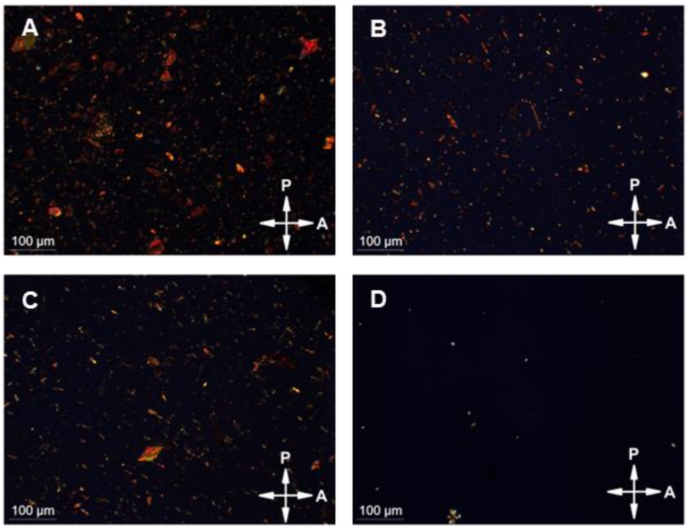

Fig. 5. POM images revealing the phase behavior of the PDLC composites
For the compositions of A, B, C, D see Table 1

3.3 Electro-optical measurements

Electro-optical (EO) responses of PDLC films are shown on Fig. 6. There are many factors that could affect the driving voltage: film thickness, liquid crystal droplet size, interaction between liquid crystal and polymer, and so on. Since the liquid crystalline films were prepared by the same method and had the same thickness, the main factors affecting the EO efficiency of LC films were the droplet size of liquid crystals and the inter-molecular interaction between liquid crystal molecules and the polymer network.

As shown in Fig. 6 a), the best EO response was recorded for sample B, which displays good switching time values at moderate driving voltages (Fig. 7). In contrast, the EO response for samples A and C is distorted and appears as a triangular waveform instead of a rectangular one, because in these cases the switching times are comparable with the period of the applied burst signal (i.e. 5 s) (Fig 6. a) samples A and C). In addition, the driving voltage for sample A is very high. Therefore, it can be considered that these two samples do not form good PDLC films. In the case of D sample, the ON state transmission is limited to 70% and the driving voltage has a rather high value (140 V). The detailed analysis of the electro-optical properties of sample B highlights the increase in ON state transmittance with the driving voltage amplitude up to 120 Vpp when the ON state transmittance is 15% higher than in the OFF state (Fig. 8.)

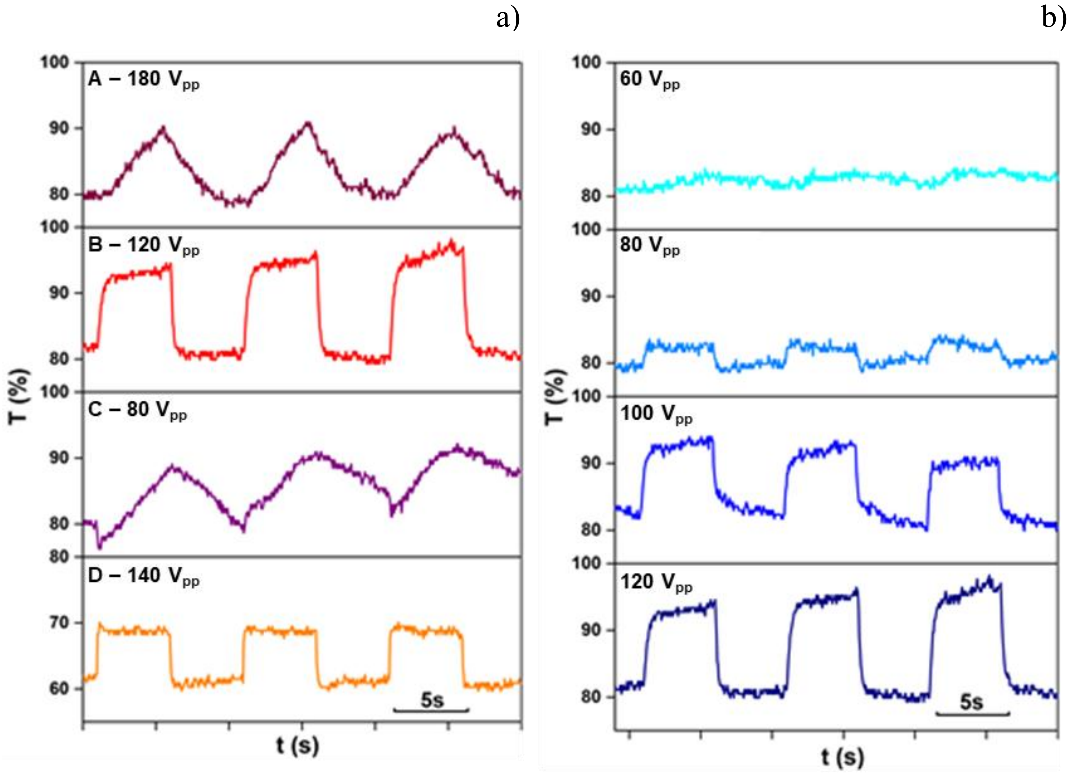

Fig. 6. a) Typical electro-optical response of PDLC cells (labeled with A, B, C, D compositions - in each case the driving voltage amplitude corresponds to the optimal response). b) The electro-optical response of cell filled with composition B with the increase in driving voltage amplitude.

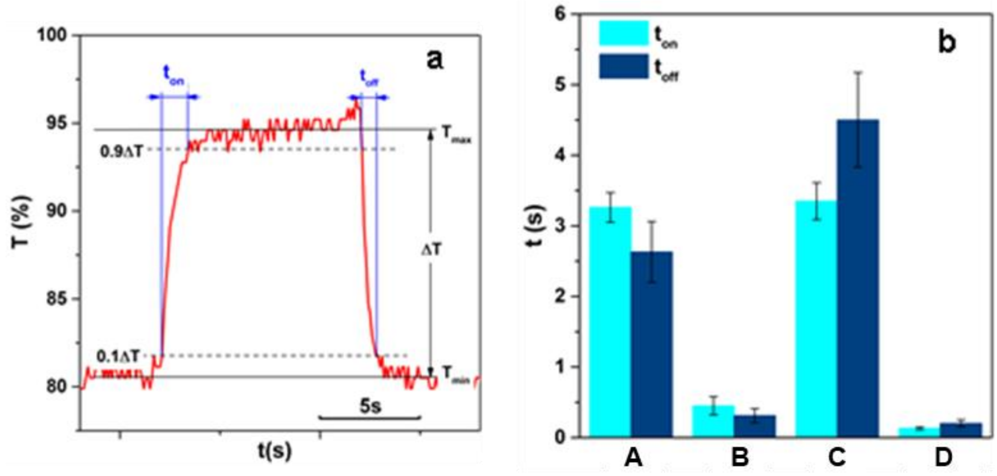

Fig. 7) a) Evaluation of switching times ($t_{on}$and t_{off}); b) $t_{on}$and t_{off} values for all samples.

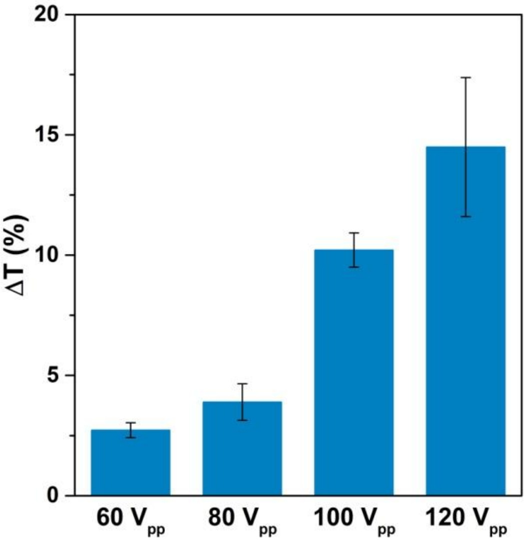

Fig. 8. Transmittance of cell II (ΔT %) depending on the V_{pp} amplitude.

This behavior can be correlated with the structure of the polymer matrix. In the case of samples A and C, the strong interaction of LC molecules with the dicyanoazobenzene sequence leads to a considerable increase in both t_{on} and t_{off} switching times. In addition, the strong anchoring phenomena to the zwitterion-doped polymer network explains the increased driving voltage value necessary for changing the orientation of LC molecules.

In sample B, the nitroazobenzene sequence, with high molar volume, favors phase separation of LC droplets. At the same time, the improved electro-optical response suggests a weaker dipole–dipole interaction between the polymer chains and LC molecules, presumably due to the lower polarizability of nitroazobenzene groups. These results confirm the existence of a correlation between the electro-optical efficiency and ground state dipole moments, to which the zwitterion substantially contributes.

4. Conclusions

Considering the fact that benzoquinonemonoimine zwitterions have stable internal dipole of 10 D, all the PDLC compositions were investigated in order to determine the influence of the polymer matrix. The study highlights the significant role of zwitterion and polymer matrix in mediating CL segregation in the composite. Among the four compositions examined, composition B containing a nitrobenzene sequence exhibits the most favorable results. The presence of the nitro moieties favors the segregation of the LC with the best electro-optical response.

Overall, this study underlines the impact of small changes upon the final properties of the PDLC materials.

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