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Thermo-mechanical and electro-optical properties of polymer dispersed liquid crystal films

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We have observed a strong dependence of the electro-optical properties of polymer dispersed liquid crystal (PDLC) films on temperature. One plausible explanation for this dependence is the thermal expansion of the films. We investigated the thermal expansion of various PDLC films using a thermal mechanical analyser (TMA). As the temperature increased, the films expanded and their droplet size decreased, and their contrast, response time, and hysteresis changed simultaneously. We observed obvious changes of thermal expansion coefficient of the films. We studied the qualitative relationship between thermo-mechanical properties and electro-optical properties. The thermal expansion of PDLC films could be adjusted by changing the degree of cross-linking of the polymer.

1. Introduction

Polymer dispersed liquid crystal films are particularly attractive for certain types of displays and light shutter applications. The absence of polarizers substantially reduces light loss and increases the brightness of both direct view and projection displays [1, 2]. The electro-optic properties of polymer dispersed liquid crystal films are highly dependent on the liquid crystal and polymer material, droplet size, shape and temperature [3-6]. Recently we studied the relationship between the morphology and the electro-optic properties of liquid crystal-polymer composite materials and found their driving voltage and transmittance to depend strongly on the polymer materials used [7]. The variations of electro-optic properties are due to the nature of liquid crystal-polymer phase separation, mode-droplet type or network type. Additionally, we observed the dependence of thermo-mechanical and electro-optical properties on temperature. The optical anisotropy of the liquid crystal reaches a maximum at low temperature, and decrease monotonically as temperature is increased [8]. The contrast decreases because the optical anisotropy of the liquid crystal is reduced at higher temperature [9]. In addition to the variation of liquid crystal properties such as viscosity, electrical and optical anisotropy, we hypothesized that these electro-optic properties are affected by the thermal expansion of the polymer. Generally polymers show thermal expansion with the increase of the temperature. Deformation of the liquid crystal droplet (or network) is due to the polymer expansion. This causes changes in

the PDLC electro-optic properties, such as operating voltage, response time, contrast, and hysteresis.

In this paper we present evidence that the changes of electro-optic properties of PDLC films are caused by their thermal expansion. We controlled the thermal expansion of PDLC by varying the degree of cross-linking of the polymer.

2. Experimental

The sample solutions were prepared by mixing the liquid crystal (80 wt %) and the acrylic prepolymers (20 wt %). The liquid crystal used was TL205 and the photoinitiator was Darocure 1173 obtained from E. Merck. The prepolymers are composed of monofunctional monomer 2-ethylhexylacrylate (EHA), aliphatic urethane-diacrylate oligomer CN963 (Sartomer Co.), and multifunctional acryl monomer pentaerythritol triacrylate (PETA), 1,6-hexandiol diacrylate (HDODA), and ethoxylated-bisphenol A diacrylate (EO-BDA). The constituents of the ultraviolet curable prepolymers are listed in the table. The mixtures are placed between indium-tin oxide coated electrodes separated by a 10 μm thick micropearl spacer, and exposed by ultraviolet (UV)

Table. The polymer constituents.

Material	Monofunctional acryl monomer	Oligomer	Multifunctional acryl monomer
S-1	EHA	CN963	PETA
S-2	EHA	CN963	HDODA
S-3	EHA	CN963	EO-BDA

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light. High pressure mercury arc lamp EN-280L from Spectronics Co. was used as the UV source.

The electro-optical measurements were described elsewhere [7]. The samples were placed in a specially designed chamber to control their temperature.

The approximate size of the liquid crystal droplets was estimated by measuring the size of the cloverleaf scattering pattern observed by laser (632.8 nm) illumination [10, 11]. Mettler FP82 HT hot stage was used to control the temperature. Optical polarizing microscope Optiphot2-POL from Nikon Co. was used.

The coefficient of thermal expansion was measured by using thermal mechanical analyser (TMA) from Polymer Laboratories Co., TMA 1000.

3. Results and discussion

Electro-optic characteristics of the various PDLC films with different types of polymer and liquid crystal mixtures are studied. Figure 1 shows the effect of monomer functionalities on the contrast-temperature behaviour. Sample S-1 which used trifunctional monomer PETA shows highest contrast. This is due to the high cross-linking density of the PETA. The difunctional monomer EO-BDA, HDODA have the fast-cure response properties rather than cross-linking. The contrast of the films initially increased with temperature, reached a maximum at 15–25°C, and then decreased at higher temperatures. The contrast increased with temperature at early stage because the droplet size decreased as the polymer expanded. As the polymers expand, droplet sizes become small and contrast increases. But as the temperature increased further, optical anisotropy of the liquid crystal decreased and this reduced the contrast [10]. Another possible explanation is that at low temperature the droplets are randomly oriented and results in a good mismatch of indices of refraction between the polymer and liquid crystal, and at elevated temperature

the symmetry axes of the droplets take on a preferred direction because the droplets become distorted by thermal expansion of polymer matrix, thus reducing their scattering power [12]. At 15–30°C, we observed with optical polarizing microscopy that the droplets size reduced and the polarizing plane of the LC changed. As a result some dark parts of the films turned to bright under cross nicole state polarizers.

We measured the droplet size variation with temperature. Figure 2 shows the effect of temperature on the droplet size of PDLC films. The liquid crystal droplets decrease to cause the polymer to expand as the temperature increases. Polymer expansions depend on the cross-linking degree of the polymer. We investigated the PDLC film expansion in detail. Figure 3 shows CTE curve of the PDLC films. The CTE of the PDLC films increase rapidly up to 25°C, then smoothly up to 50°C. Also, highly cross-linkable monomer PETA blended sample S-1 has the lowest CTE value. The variations of contrast depend mainly on the variation of CTE trends. From these results, it is better to change the onset temperature of CTE variation of the films in order to control the electro-optical property of the film. Figure 3 also shows that polymer expansion leads to variation of contrast at low temperatures.

Response time, especially decaying time is dependent on the droplet size, shape and liquid crystal viscosity, and elastic constant [13, 14]. West *et al.* studied unusual electro-optic effects at elevated temperatures to changes in the surface anchoring strength as a function of temperature, as well as the variations in the dielectric properties and droplet shape [15]. We have investigated response time and hysteresis variation. Figure 4 shows the variation of decay time with temperature. Initially (10–30°C) the decay time decreases due mainly to the decrease of droplet size, while at high temperature (above 30°C) the decrease

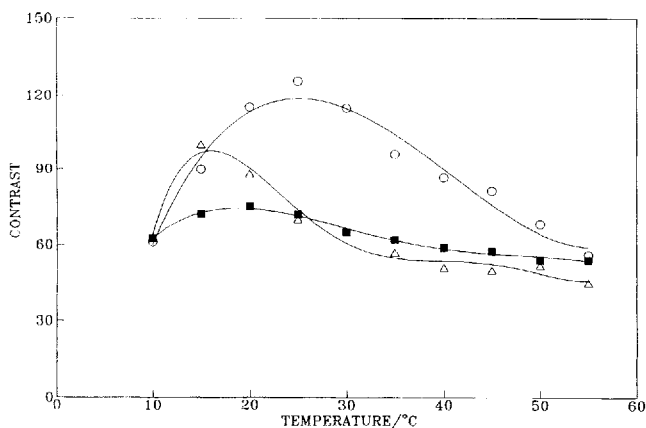


Figure 1. The effect of the temperature on the contrast of the PDLC films; S-1 (○), S-2 (■) and S-3 (△).

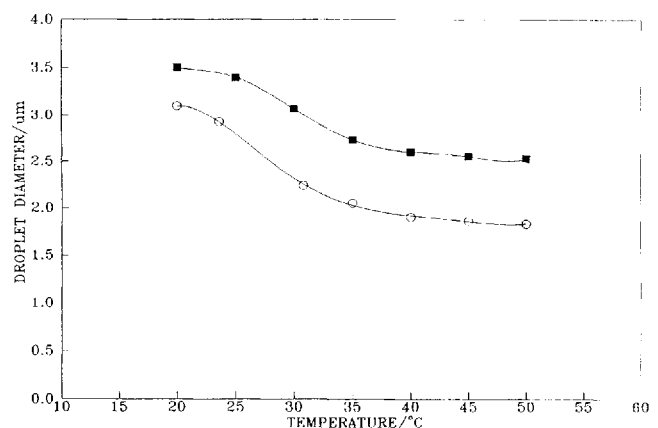


Figure 2. The effect of the temperature on the liquid crystal droplet diameter of the PDLC films; S-1 (○) and S-2 (■).

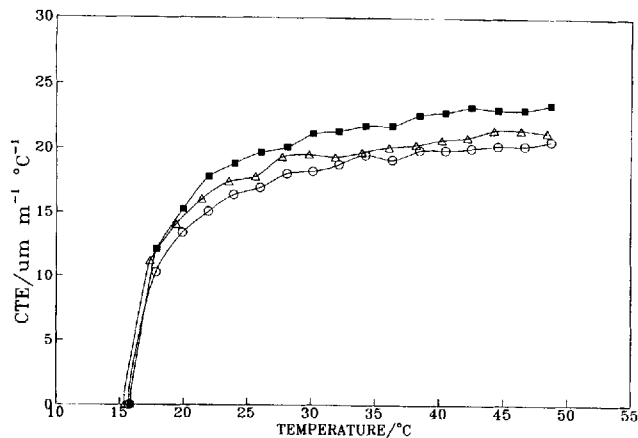


Figure 3. The effect of the temperature on the coefficient of thermal expansion of the PDLC films; S-1 (○), S-2 (■) and S-3 (△).

is mainly due to viscosity and surface tension. Considering the decrease of decay time at 10–30°C, droplet size variation mainly leads the response property of PDLC films below 30°C. As the temperature was increased, the polymer expanded and the droplet size reached its minimum because liquid crystals in droplets resist further shrinkage.

Figure 5 shows the hysteresis of the films with temperature. The films show different hysteresis behaviour below 25°C. The hysteresis of the films decrease rapidly at 10–25°C, while there is no variation above 25°C. This is the behaviour one typically imagines: by thermal expansion of the polymer, the liquid crystal droplets became smaller and distorted. Simply shearing the PDLC films caused the decaying time to drop from >100 ms to <5 ms [14]. Hysteresis, to some extent, decreased by shearing. The liquid crystal droplets that were strongly distorted showed a small hysteresis [16, 17]. The abrupt changes in the electro-optical properties of the

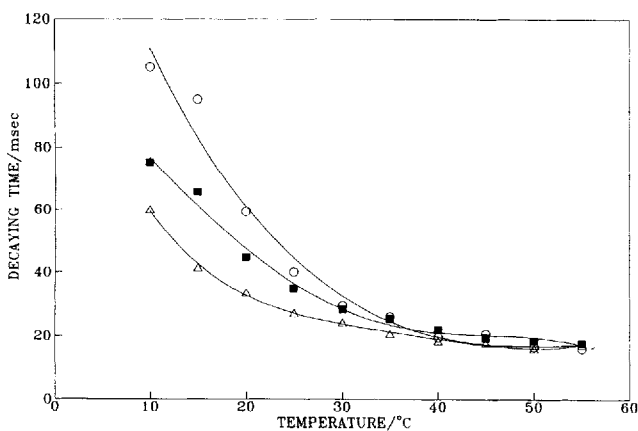


Figure 4. The effect of the temperature on the decay times of the PDLC films; S-1 (○), S-2 (■) and S-3 (△).

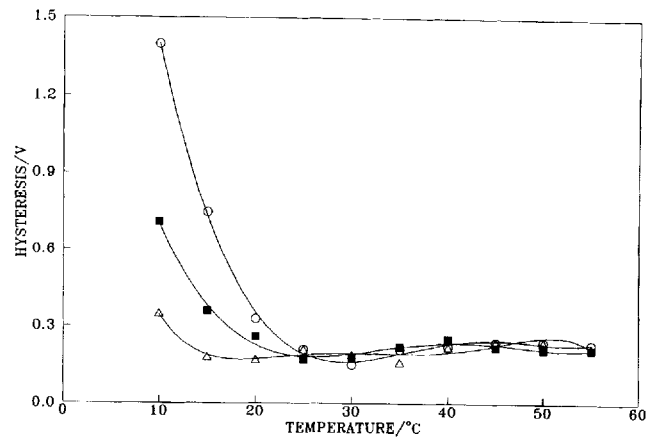


Figure 5. The effect of the temperature on the hysteresis for the PDLC films; S-1 (○), S-2 (■) and S-3 (△).

PDLC films were affected by the changes at the liquid crystal/polymer interface at the glass transition temperature of the polymer [15]. It is very interesting to note that the decrease of response time and hysteresis is coincident at 10–25°C. This means that droplet size decreases and droplet distortion causes the polymer to expand below 25–30°C, while above this temperature droplet configuration reached a certain stable state. The variations of electro-optic properties such as decay time and hysteresis are well correlated with that of thermo-mechanical properties of the polymer at 15–25°C. From these results we can make an analogy between polymer expansion, response times and hysteresis decrease. Thermal expansion of the polymer dispersed liquid crystal reduced the droplet size, distorted the droplet shape, and as a result affected contrast, response times, and hysteresis. By selecting appropriate monomers, we can adjust polymer expansion and the electro-optic properties of the PDLC.

4. Conclusions

We have shown that the electro-optical properties are well correlated with the thermo-mechanical properties of the PDLC films. Electro-optical properties depend not only on physical properties of the liquid crystal but also on the thermo-mechanical properties of the polymers. As the polymer expands, the liquid crystal droplet size reduced and shape distorted. As a result, contrast, decay time, and hysteresis varied with temperature. We used various polymers to study the effect of thermal expansion on the electro-optic properties. PDLC films made with a highly cross-linkable trifunctional monomer showed the highest contrast, and lowest thermal expansion.

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