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Polymer-dispersed liquid crystals: Preparation, operation and application

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Invited Article

Polymer-dispersed liquid crystals: Preparation, operation and application

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Liquid crystal–polymer composites have been widely studied in the last ten years, both because of potential applications and to improve our understanding of the behavioural mechanisms involved. Although it cannot be exhaustive, this review describes recent advances in the field of liquid crystal–polymer composites. The preparation techniques as well as the operating principle and various applications of these materials are covered.

1. Introduction

Up to now, owing to its unique features, the twisted nematic (TN) effect is the most widely used electro-optical effect for making flat panel displays. Improvement of liquid crystal materials in conjunction with all the aspects of display technology (alignment layers, microlithography, thin film transistors, etc.) made possible the rapid progress which led within twenty years from simple digital watch displays to high information content liquid crystal displays (LCDs). Nevertheless, the TN effect suffers from some basic drawbacks (poor viewing angle, need of polarizers and alignment layers) which limit growth in its applications. The strong demand for large area, direct view and projection LCDs able to reproduce high definition coloured pictures for personal computer terminals, television screens and large area information boards, requires the development of new electro-optical materials and/or new electro-optical effects. In the mid 1980s a novel class of electro-optical materials was introduced. They consist, in their most common form, of low molecular weight nematic liquid crystalline microdroplets dispersed in a solid polymer matrix. These liquid crystal polymer composites (LCPCs) exhibit some specific properties that make them very promising for display device applications. Now, several types of LCPC differing in their polymer content or the nature of the liquid crystalline phase have been prepared and evaluated, and many potential applications have been demonstrated. A thorough review of the enormous amount of literature published within these last ten years would be beyond the scope of this paper. But in the following, the main aspects of the electro-optical properties of LCPCs, as well as the different preparation methods and the wide variety of potential applications, will be presented and discussed.

2. Historical background

The possibility of changing a liquid crystal composite from an opaque to a transparent state was first demonstrated by C. Hilsum in 1976 [1], with micron sized glass beads dispersed in a nematic liquid crystal. Later, H. G. Craighead [2] applied the same principle with a porous polymer matrix (a filter of esters of cellulose) filled with a liquid crystal. Probably due to the poor scattering of these two configurations, they were not developed further, but gave rise to two successful approaches. First in 1985, J. L. Fergason [3] and P. S. Drzaic [4] reported liquid crystal–polymer composites obtained by drying an emulsion of a liquid crystal in an aqueous solution of polyvinyl alcohol. These materials, called NCAP for nematic curvilinear aligned phase, were soon developed for smart window applications. A year later, J. W. Doane introduced a second class of composites called PDLC (polymer-dispersed liquid crystal) by inducing phase separation in a homogeneous mixture of a liquid crystal and an epoxy prepolymer [5].

Since then, a tremendous activity has characterized this field of research, ever increasing the scope of the applications of these materials. Several reviews have

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been published [6, 7, 8], and the aim of the present review is to update them with a comparison between theoretical models and recent experimental results. In addition, applications of liquid crystal–polymer composites are examined.

3. Electro-optical properties

The liquid crystal employed can be nematic as well as cholesteric or smectic, but nematic liquid crystal–polymer composites are by far the most studied. Moreover, because several preparation methods exist and because of the wide variety of constituents, composites can have very different microscopic structures. Some composites are really a dispersion of droplets of liquid crystal in a polymer film, but it is also possible to obtain a network of polymer in a continuous liquid crystalline medium. Nevertheless, the electro-optical effects of all these composites are similar.

3.1. *Principle of operation*

A composite consists of a film of polymer in which randomly oriented liquid crystal domains are dispersed (figure 1). Because of refractive index variations, the film is scattering. If transparent electrodes are applied on each side of the film, the nematic liquid crystal domains can be oriented by an electric field, and if the ordinary refractive index of the liquid crystal is adjusted to the index of the polymer, then the film becomes transparent. When the electric field is removed, the liquid crystal domains recover their initial orientations because of surface forces and because the cavities are not ideal spheres.

3.2. *Advantages over other technologies*

In comparison with existing display technologies, liquid crystal–polymer composites are potentially attractive for several reasons. First, the on-state is close to one hundred per cent transparent because no polarizer is present. Second, these composites can easily be processed because of the polymer binder, and finally, no surface treatment is required, eliminating a technological drawback of twisted nematic liquid crystal displays.

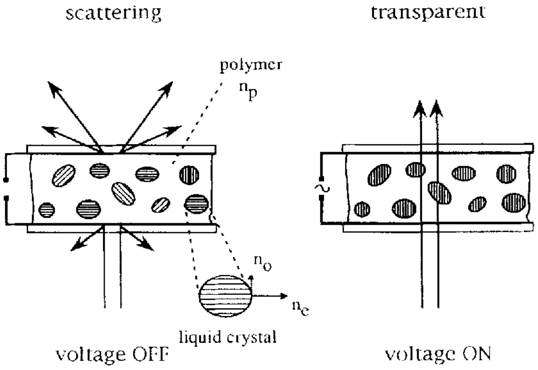

Figure 1. Liquid crystal–polymer composite: principle of the electro-optical effect.

3.3. *Properties of liquid crystal–polymer composites*

It is possible to characterize a liquid crystal–polymer composite film by measuring the optical transmission of a laser beam through the film with a photodiode placed behind a diaphragm. This optical transmission is recorded as a function of the voltage applied to the film, and can be represented as in figure 2, where several properties are indicated: minimum transmission $T_{\min}$; maximum transmissition $T_{\max}$; V_{10}, V_{90} and V_{sat} are the voltages necessary to obtain a transmission of 10%, 90% and 100%, respectively; hysteresis (ΔV_{50}) between the electro-optical curves obtained by increasing and decreasing the voltage.

3.3.1. *Contrast*

3.3.1.1. *Definition.*

Contrast is a very important property of composites, but it cannot be easily compared in the literature. Indeed, several definitions of contrast are used [9], although the most widespread is: contrast $= T_{\max}/T_{\min}$.

More insidious is the very large dependence of contrast values on experimental conditions such as: the nature of the light source (collimated beam, diffusive source, etc.); the wavelength of the light source ($T_{\min}$ can vary by a factor of two between 400 and 680 nm [10]); the collection angle of the photodiode. Unless these experimental conditions are specified, it is not possible to compare two values of contrast.

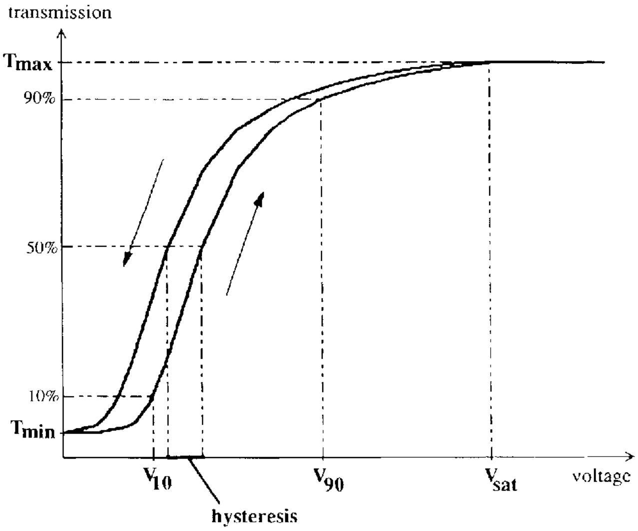

Figure 2. Electro-optical curve of a liquid crystal–polymer composite.

3.3.1.2. *Theoretical results.*

Even if it is easy to understand why a liquid crystal–polymer composite is scattering, it is not straightforward to quantify this scattering. Some authors have tried to tackle this subject, but not very successfully, because it is so complex that too many simplifications are required [11]. In fact, to answer this problem, one needs to be able to calculate the trajectory of a light beam through a film containing a large number of anisotropic droplets whose refractive indices are not uniform within each droplet, whose axes of symmetry are randomly oriented and whose sizes are not constant throughout the film.

Nevertheless, G. P. Montgomery *et al.* [12] have applied Rayleigh–Gans theory to show that if liquid crystal anisotropy can be neglected and if the droplet density is low enough so that each light beam is scattered only once in the film, then back scattering is a maximum for a droplet diameter equal to $\lambda/7$, where λ is the wavelength of the light source. Others have shown that the anomalous diffraction model can be applied to systems with a low density of large droplets (more than $20\,\mu m$ [13] or $1\,\mu m$ [14] if refractive index differences are low). On the other hand, J. R. Kelly *et al.* [15] have stressed that multiple scattering cannot usually be neglected.

Even if a quantitative description of scattering is not feasible at the moment, it is possible to deduce a few qualitative rules from experimental results.

3.3.1.3. *Experimental results.*

Film thickness. The thicker the film, the more scattering it is in the off-state, and this dependence is close to an exponential law [16–19]. On the other hand, on-state transparency is nearly independent of thickness, as long as it is possible to apply a sufficiently strong voltage.

Droplet diameter. Very large and very small droplets do not scatter light efficiently, so that there is an optimum size which seems to be close to $5 \times \lambda$, i.e. close to $3\,\mu m$ [20–23].

Liquid crystal content. Similarly, a very large or very low proportion of liquid crystal does not scatter efficiently. The optimum depends on the particular nature of the composite, but several authors have reported that this optimum is in the range of 60 weight per cent of liquid crystal for their systems [24, 25].

Refractive indices. Off-state scattering is a function of refractive index differences: between a liquid crystal droplet and the polymer [26]; between two adjacent droplets [16, 27]; within a droplet of liquid crystal [26, 27].

Consequently, whatever the main scattering mechanism, increasing the liquid crystal refractive index anisotropy enhances off-state scattering [28, 29]. This can be achieved with liquid crystal mixtures containing highly polarizable molecules such as cyanoterphenyls [30, 31]. In order to have a highly transparent on-state, the polymer refractive index (n_p) must be adjusted to the ordinary refractive index of the liquid crystal (n_o) [32].

Solubility. Adjusting refractive indices is not straightforward, because they are a function of inter-solubility of the polymer and the liquid crystal [33]. As much as 30% of the liquid crystal can be dissolved in the polymer binder [34], even if the latter is crosslinked [35]. Because of the liquid crystal, the polymer index in the composite (n'_p) is increased, so that it is necessary to choose a polymer whose index (n_p) is slightly lower than n_o: $n_p < n'_p = n_0$ [29, 36].

Moreover, gas chromatography of the liquid crystal contained inside the droplets of a composite has proved that the different constituents of the liquid crystal mixture do not have the same solubility [37]. Lower molecular weight components are more soluble in the polymer, so that the composition and thus the properties of the liquid crystal mixture are altered. Finally, oligomers from the binder can remain dissolved in the liquid crystal.

3.3.1.4. *Viewing angle.*

We have mentioned that equality between refractive index of the polymer inside the composite (n'_p) and n_o provides a highly transparent on-state, but that is only true for a light beam perpendicular to the film. If the incidence angle is not zero, then the effective refractive index of the liquid crystal in the on-state is between n_o and n_e, so that the film is hazy. In order to reduce this haze at large viewing angle, it is possible to choose a polymer and a liquid crystal with $n'_p > n_0$, so that indices are adjusted for an oblique angle [38].

Another possibility for improving viewing angle, but only in one direction, consists in placing a polarizer in front of the liquid crystal–polymer composite [39]. Finally, a polymer liquid crystal constitutes an anisotropic binder, in which both ordinary and extraordinary refractive indices can be adjusted, in order to suppress haze. This type of composite is examined in § 6.1.

3.3.1.5. *Influence of temperature on contrast.*

Polymer refractive index is a slowly decreasing function of temperature, but n_o increases and n_e decreases when the temperature is increased, and these variations are very significant close to the clearing point T_{N-I} (figure 3) [32]. Consequently, both $(n_e - n_p)$ and $(n_e - n_o)$ are reduced at higher temperature, so that off-state scattering is reduced [18, 40]. On the other hand, a temperature can exist at which $n_p = n_o$, so that on-state transmission goes through a maximum [41]. Furthermore, temperature can modify the solubility of

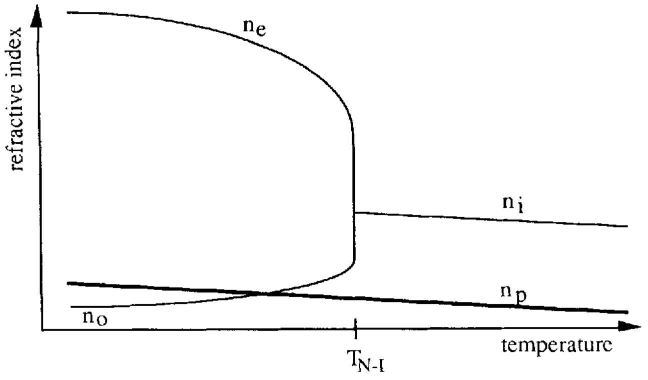

Figure 3. Qualitative temperature dependence of refractive indices of the polymer (n_p) and the liquid crystal (n_o = ordinary, n_e = extraordinary, n_i = isotropic refractive index).

the liquid crystal in the polymer [33] and thus the refractive indices. Nevertheless, these variations can be avoided if a high T_{N-I} liquid crystal is used [29].

3.3.2. *Driving voltage*

3.3.2.1. *Definitions.*

Driving voltage is generally chosen as V_{90} (figure 2), but can also be V_{sat}. Threshold voltage is usually defined as V_{10}. All these values depend on T_{min} and T_{max}, and as a consequence, they are also a function of measuring conditions (such as light source, wavelength, collection angle, etc.) [10], as well as frequency and shape of the electric signal.

3.3.2.2. *Theoretical results.*

Director field without an electric field. In a cavity, the liquid crystal is submitted to surface forces, so that the orientation of the director is imposed by the shape of the cavity, the interaction between the polymer and the liquid crystal and the elasticity of the liquid crystal. In the case of spherical cavities, the director field can be calculated by minimizing the free energy of a droplet, composed of a term of elastic deformation and a term of anchoring [6, 42–44]. The configuration of the director depends on anchoring conditions. In the case of homeotropic anchoring, two stable configurations have been identified: the radial configuration, with a spherical symmetry and a point defect in the middle; the axial configuration, with a cylindrical symmetry and an equatorial line defect. In the case of planar anchoring, mainly three other configurations are stable: the bipolar configuration, with a cylindrical symmetry and two point defects (the poles); the concentric configuration, with a cylindrical symmetry and a line defect in the middle; the twisted bipolar configuration [43(*b*)], which is intermediate between the last two.

These theoretical configurations have been observed experimentally by several techniques. Polarizing optical microscopy is the most direct method, and enables observation of liquid crystal droplets dispersed in a solid polymer [45], as well as in a liquid (glycerol [46], polydimethylsiloxane [47]). Pictures are interpreted with the help of numerical simulations [45, 48–50]. However, this method is limited to a low concentration of droplets and to droplets larger than ten microns. It is difficult to apply these results to smaller droplets, because the configuration of a liquid crystal droplet in a particular polymer depends on the size of the droplet.

Infrared micro-spectroscopy has also been used, but it suffers from the same limitations [51]. Deuterium NMR is a useful tool for the determination of the configuration of small droplets ($<1\,\mu$m) which are not oriented by the magnetic field [52, 53]. In that case, it is necessary to use a partly deuteriated liquid crystal, but solid state NMR can determine if the anchoring is homeotropic or planar without deuteriation [54].

Liquid crystal response under an electric field. The electrical response of a composite has been studied [55, 56], but the liquid crystal response has been described only for two cases: the radial and bipolar droplets. Application of an electric field transforms a radial configuration into an axial one. Evaluation of the energy difference between these two configurations yields the minimum voltage required to switch the liquid crystal [6, 44, 57]:

$$V_R = c\left(\frac{\rho_p}{\rho_{lc}}+2\right)\frac{dW_0}{(K\varepsilon_0\Delta\varepsilon)^{1/2}} \qquad (1)$$

where d is the thickness of the film, W_0 the anchoring energy, K the elastic constant, ε_0 the dielectric permittivity, $\Delta\varepsilon$ the dielectric anisotropy of the liquid crystal, ρ_p and ρ_{lc} the resistivities of the polymer and the liquid crystal, respectively, and c is a numerical constant. Thus the driving voltage appears to be an increasing function of film thickness, anchoring energy, and resistivity ratio, but a decreasing function of elasticity and dielectric anisotropy of the liquid crystal.

In the more frequent case of a bipolar configuration, inside an elongated droplet whose long axis is in the plane of the film, the driving voltage has also been calculated:

$$V_B = \frac{d}{3a}\left(\frac{\rho_p}{\rho_{lc}}+2\right)\left(\frac{K(l^2-1)}{\varepsilon_0\Delta\varepsilon}\right)^{1/2} \qquad (2)$$

with $l = a/b$, where a and b are the long and short axes, respectively, of the ellipsoid [6, 58]. To be able to derive this expression, the authors have neglected anchoring forces compared to the shape anisotropy of the droplet. Except for liquid crystal elasticity, the influence of the other parameters is the same as in the radial case.

3.3.2.3. *Experimental results.*

Comparison between theoretical and experimental results found in the literature can only be qualitative because experimental conditions are seldom identical.

Film thickness. The linear dependence between film thickness and driving voltage predicted by equations (1) and (2) is usually verified [28, 59]. However, in some cases the driving voltage increases if the film thickness becomes very low ($<10\,\mu m$) [17]. This behaviour could be due to the larger fraction of droplets that are in contact with the electrodes and are consequently deformed.

Droplet diameter. According to equation (2), driving voltage should be inversely proportional to droplet diameter. This relationship is verified by several experimental results [60, 61]. Other materials show a more complex dependency between driving voltage and droplet diameter, although it is still a decreasing one [20, 59]. Furthermore, S. Tahata *et al.* [23] show that for a large diameter ($>3\,\mu m$), the driving voltage becomes independent of diameter, and H. Nomura *et al.* [21] report that for droplets larger than $4\,\mu m$, the driving voltage increases! These last authors attribute this phenomenon to an increase in the average dielectric constant when a droplet becomes larger. Another explanation is that the size of these droplets becomes close to the film thickness ($10\,\mu m$), so that more and more droplets are deformed by the electrodes. Thus it can be concluded that driving voltage is a decreasing function of droplet diameter, as long as the latter is small compared to film thickness.

Droplet shape. For elongated droplets having their long axes in the plane of the film, equation (2) predicts a large increase in driving voltage with droplet eccentricity. J. H. Erdmann *et al.* [44] have shown that driving voltage is reduced for elongated droplets tilted by an angle of 56° to the normal to the film, compared to spherical droplets. However, this result does not contradict the model because, in the elongated case, the liquid crystal has to be rotated by 56° and not up to 90° as in the case of spherical droplets.

Elastic constant and dielectric anisotropy of the liquid crystal. We were not able to find any direct study of the influence of liquid crystal elasticity and dielectric anisotropy. However, since these parameters are independent of the polymer, it can be assumed, as a first approximation, that driving voltage is an increasing function of K and a decreasing one of $\Delta\varepsilon$, as in the case of pure nematic liquid crystals in cells [62].

Resistivities. According to equation (2), driving voltage should be a linear function of ρ_p/ρ_{lc}, but this is difficult to check because resistivities of the polymer and liquid crystal in the composite can be very different from those of the pure materials [63]. Nevertheless, J. H. Erdmann *et al.* [44] have adjusted the crosslinking density of their epoxy-based PDLCs, in order to control the resistivity of the binder. They have reported a linear dependence between driving voltage and resistivity, but this approach neglects the effect of changing the crosslinker content on the chemical nature of polymer chain ends and thus on anchoring energy.

In the case of NCAP materials based on polyvinyl alcohol, L. Welsh *et al.* [64] showed that even though purification by Soxhlet extraction with ethanol increased the resistivity of the polymer by two orders of magnitude, they noticed no evolution of the electro-optical curve. Furthermore, A. Seeboth [65] showed that the driving voltage of the composite is decreased if a salt (NaCl) is added to the gelatine matrix, but the effect on ρ_p/ρ_{lc} is not reported. Finally, the influence of resistivity is frequency dependent: at low frequency ($<100\,Hz$), ionic impurities shield the local electric field, so that it is necessary to apply a larger voltage to drive the composite [66–69]. It seems that the optimum frequency is in the kHz range, because at high frequency, dielectric dissipation increases [70].

Anchoring. Anchoring of a liquid crystal on a flat surface is still a subject of controversy [71–73] and anchoring at the surface of droplets is even more complex [74, 75]. Several publications show indirectly the importance of anchoring. For instance, C. A. McFarland *et al.* [76] have shown that a liquid crystal is homeotropically oriented in a composite made from poly-*iso*butyl methacrylate, but that it is planar if the polymer is poly-*n*-butyl methacrylate. Another example is the composites made from gelatine: the orientation of the liquid crystal droplets depends on the organization of the polymer [77, 78]. Finally, the existence of the so called 'memory effect' (see § 5.3.4) also shows the importance of anchoring of the liquid crystal [79].

Consequently, several studies on model systems have been reported [80–82], but it is difficult to relate these results to electro-optical properties of liquid crystal–polymer composites. At the same time, some research groups have modified the chemical composition of the polymer in the composite and derived conclusions about anchoring [59, 83–92]. Unfortunately, in these studies, it is impossible to really distinguish the effect of anchoring on electro-optical properties, because other parameters such as refractive index, resistivity or droplet size and shape are not constant. Additional experiments on these systems involving second harmonic generation measurements [93, 94] might give an answer to the problem.

To our knowledge, only two studies show a quantitative effect of anchoring on electro-optical properties. First H. Nomura *et al.* [95] have studied a composite whose matrix is made from micro-beads coated with a thin film ($0{\cdot}1\,\mu m$) of a copolymer of methyl methacrylate and (methoxypolyoxyethylene) methacrylate. Increasing the content of methyl methacrylate in the copolymer leads to an increase of driving voltage, so that anchoring is stronger on poly(methyl methacrylate) than on poly(methoxypolyoxyethylene) methacrylate. The second study [96] consists of comparing electro-optical properties of composites where only the surfaces of the pores in the polymer binder have been chemically modified. These results show that for a cyanobiphenyl liquid crystal, fluorinated or hydroxylated chemical moieties on the surface of the polymer induce a stronger anchoring than alkyl moieties. This phenomenon seems to be due to the enhanced compatibility between the liquid crystal and the polymer surface, in the case of alkyl moieties.

3.3.2.4. *Influence of temperature on driving voltage.*

When temperature is increased, driving voltage is usually reported to decrease [24, 44, 97, 98], and then finally to increase [29] if the clearing point is approached. The decrease is due to a lower $K/\Delta\varepsilon$ [41] and to lower anchoring forces. This last point has been shown by solid state NMR experiments proving that the contact time of mesogens on the surface of the polymer decreases with temperature [54]. On the other hand, resistivity of the liquid crystal decreases more rapidly than resistivity of the polymer, so that ρ_p/ρ_{lc} increases [68]. This could explain the high temperature increase of the driving voltage, the more so as electric field shielding by ions is favoured [69]. Moreover, heating the composite at a temperature higher than the glass transition temperature of the polymer [75, 99], or the effect of increased inter-solubility between the polymer and the liquid crystal [33] can modify the general pattern.

3.3.3. *Response times*

3.3.3.1. *Definition.*

Rise time (τ_{on}) is usually defined as the time needed for the composite to reach 90% of the on-state transmission when an alternating voltage of amplitude V is applied. Similarly, decay time (τ_{off}) is the time needed for a composite in the on-state to reach 10% of the on-state transmission when the voltage is set to zero. To a first approximation, decay time does not depend on the measuring voltage V, but rise time is inversely proportional to V^2 [18, 100]. As a result, comparison of rise time values in the literature is only possible if measuring voltages are the same, which is seldom the case.

3.3.3.2. *Theoretical results.*

In the case of elongated droplets of bipolar configuration whose long axes are in the plane of the film and if liquid crystal anchoring is neglected, it is possible to obtain expressions of response times [6, 44]:

$$\tau_{on} = \gamma\left[\frac{9\varepsilon_0\Delta\varepsilon V^2}{d^2[2+(\rho_p/\rho_{lc})]^2} + \frac{K(l^2-1)}{a^2}\right]^{-1} \approx \gamma\left(\frac{d}{V}\right)^2 \frac{[2+(\rho_p/\rho_{lc})]^2}{9\varepsilon_0\Delta\varepsilon} \tag{3}$$

$$\tau_{off} = \frac{\gamma a^2}{K(l^2-1)} \tag{4}$$

where γ is the rotational viscosity of the liquid crystal.

Rise time is mainly a function of electric field (V/d), viscosity, resistivities and dielectric anisotropy. Decay time depends on viscosity and elasticity of the liquid crystal, as well as on shape and size of the droplet.

3.3.3.3. *Experimental results.*

It is possible to compare qualitatively these theoretical expressions with experimental results in the literature. It must be noted first that these response times are relevant to fully on and fully off states; response times between grey levels are longer [101]. Secondly, this model describes the movement of the liquid crystal droplet as one collective movement, but it has been shown that the liquid crystal can also reorient itself according to two different motions corresponding to the centre and the boundary of the droplet [102–104].

Film thickness. Of course, the speed at which the liquid crystal moves does not depend on the thickness of the film, but response times do. A thick film is more scattering in the off-state, but nearly as transparent as a thin film on the on-state. Consequently, the thick film reaches 10% of its on-state transmission more quickly; decay time is a decreasing function of film thickness [17]. Similarly, rise time is an increasing function of film thickness (at constant measuring voltage) [18].

Droplet diameter. Increasing droplet diameter reduces the magnitude of surface effects compared with bulk effects, so that decay time is expected to increase and rise time to decrease. This trend has been experimentally confirmed for decay time [20, 61], but we were unable to find clear data concerning rise time.

Droplet shape. According to expressions (3) and (4), a higher aspect ratio for the droplets should lead to a large decrease of τ_{off} and a small decrease of τ_{on}. These behaviours have indeed been observed [44, 105].

Elastic constant and dielectric anisotropy of liquid crystal. No data concerning the influence of elasticity and dielectric anisotropy on response times has been found, but Z. J. Lu *et al.* [106] have demonstrated that introducing a chiral dopant into the liquid crystal results in a decrease of relaxation time. Moreover, several authors have shown [83, 107] that decay time can be reduced significantly by addressing at high frequency a composite containing a liquid crystal whose dielectric anisotropy is positive at low frequency and negative at high frequency.

Viscosity. It seems natural that a higher viscosity of the liquid crystal should increase both response times (expressions (3) and (4)), but the true viscosity of the liquid crystal inside the composite is difficult to evaluate and it depends a lot on the nature of the polymer [29], because of dissolved oligomers.

Resistivities. The influence of resistivities on rise time is expected to be the same as on driving voltage. A. Miyamoto *et al.* [108] have indeed shown that τ_{on} is larger for a composite based on poly(methyl methacrylate) than on poly(butadiene-co-acrylonitrile) which is less resistive.

Anchoring. Decay time has been shown to increase moderately while rise time is dramatically reduced by a reduction of anchoring [96]. Similarly, B. G. Wu *et al.* [109] have shown that a small amount of polymer forming a web-like network in the liquid crystal droplets reduces decay time, probably through an increase in anchoring.

3.3.3.4. *Influence of temperature on response times.*

Rise time. Numerous experimental results show that increasing temperature reduces rise time [18, 24, 29, 41, 44, 110], probably through a decrease in liquid crystal viscosity. However, this is only true if the driving frequency is high enough, because resistivities decrease with temperature, so that the frequency below which electric field shielding occurs increases with temperature. Consequently, if the driving frequency is low, electric field shielding can be more important than the reduction in viscosity, and τ_{on} can show an increase [108].

Decay time. The effect on decay time is less clear cut: τ_{off} has been observed to decrease [41], increase [18, 24, 99] or go through a minimum [29, 44, 58, 110] when temperature is increased. It seems that, depending on the large variety of materials used, the decrease in viscosity can be cancelled by the decrease in elastic constants and anchoring forces, which tend to increase decay time.

3.3.4. *Hysteresis*

3.3.4.1. *Definition.*

Electro-optical curves measured while increasing or decreasing the voltage are generally not identical. This hysteresis is usually quantified by $\Delta V_{50} = V_{50^+} - V_{50^-}$, which measures the difference between the voltages necessary to obtain 50% of maximum transmission (see figure 2). As well as other properties, hysteresis depends on measuring conditions (light source, wavelength, collection angle, electric signal shape and frequency, etc.), but it is especially affected by the speed at which voltage is increased and decreased. Hysteresis is usually considered as a nuisance because it makes grey levels difficult to obtain [97, 101], but in some cases, it may be interesting to enhance hysteresis for memory effects [28] or bistability [111].

3.3.4.2. *Interpretation of hysteresis.*

The phenomenon of hysteresis has not yet been understood, but Z. Li *et al.* [112] have shown that hysteresis is identical whether the composite is driven by an electric or a magnetic field. This proves that hysteresis is not the consequence of charge build up or of a polarization effect. P. S. Drzaic [104] has suggested that hysteresis is due to a thin layer of liquid crystal, at the interface with the polymer, which is not oriented by the field in the same manner as the bulk in the droplets.

3.3.4.3. *Experimental results.*

Hysteresis depends mainly on the nature of the polymer and the liquid crystal, and on the preparation of the composite, but at the moment no general rules have been devised. It seems that flattening liquid crystal droplets in the film reduces hysteresis [97, 101], maybe because it increases elastic forces. It is also possible to reduce hysteresis with a dual frequency addressing scheme [83], if the sign of the liquid crystal dielectric anisotropy changes with frequency.

3.3.5. *Holding ratio*

Driving voltage is an important feature of liquid crystal–polymer composites, but it is vital that this voltage should not decrease too rapidly when the electric circuit is opened. In a display operating at video frequency, each line is addressed for a short period of time (about 60 µs), but not addressed during about 40 ms. The dielectric losses of the film must be as low as possible, so that the voltage applied during 60 µs remains for 40 ms. This property is measured by the holding ratio [113]. Of course, holding ratio depends on the nature [114] and purity [64] of the starting materials. That is why cyanobiphenyls are more and more replaced by halogenated compounds [31]. But holding ratio is also very sensitive to impurities introduced during processing, so that it is not possible to estimate the holding ratio of the composite from those of its constituents. For instance, the holding ratio of a composite made by emulsion has been reported [113] to be superior to the holding ratio of both liquid crystal and polymer!

3.3.6. *Ageing*

Stability of electro-optical properties over time is an essential property which can be assessed by several specific tests. The simplest test consists of regularly measuring the properties of a composite left at room temperature and in the off-state. Depending on the materials, large evolutions can be measured within months or even days [92]. These variations can be due to slow crosslinking processes [115] or to diffusion processes driven by thermodynamic equilibrium. They can be reduced by using high yield reactions and by increasing crosslinking density [60].

Stability can also be tested by heating a composite in a humid atmosphere: the liquid crystal seems to be well protected by the polymer if it is hydrophobic, even without sealing [116]. Photochemical stability can be evaluated by measuring the evolution of properties after irradiation at selected wavelengths. A. M. Lackner *et al.* [116] have shown that their composite is not affected by wavelengths longer than 372 nm, but that it deteriorates at 351 nm, probably because of liquid crystal destruction [30]. Finally, electro-optical properties can evolve significantly after long periods of operation at high voltages, but this point has not yet been reported.

4. Preparation

Liquid crystal–polymer composites can be prepared by several techniques which are usually described as emulsion or phase separation processes.

4.1. *Emulsion*

4.1.1. *Description*

Composites obtained by emulsion are traditionally called NCAP (for nematic curvilinear aligned phase). A liquid crystal is emulsified in an aqueous medium which contains either a water soluble polymer [117,118] or a latex [119] and usually a surfactant. The emulsion is coated on a substrate and dried. A second substrate is then laminated on top of the film, sometimes with the help of a thermoplastic sub-layer to improve adhesion [119]. Liquid crystal droplets in the film are usually flattened, first because of shrinking of the film during drying and also because of laminating [120].

4.1.2. *Advantages and drawbacks*

The main advantage of this method is its easy extension to large areas and industrial processes. Moreover, very different kinds of polymers can be used. Hydrophilic polymers (such as polyvinyl alcohol) are potentially interesting because they are not plasticised by the liquid crystal, so that all the liquid crystal is recovered in the droplets. Of course hydrophilic polymers must be carefully purified from ionic impurities [64] and thoroughly dried after coating. But it is also possible to use hydrophobic polymer dispersions (such as polyurethane or polyacrylate) which can be crosslinked. To reduce plasticising, a small quantity of hydrophilic polymer (polyvinylpyrrolidone) can be used as a barrier [119]. Size of liquid crystal droplets in the emulsion and in the dried film can easily be measured [121] and modified by adjusting surfactants or by adding an alcohol in water to reduce interfacial tension [91, 122].

The main disadvantage of the method is the difficulty of thoroughly removing water which reduces holding ratio and impairs long time ageing. Moreover, it seems that hydrophilic polymers lead to high anchoring forces and thus to high driving voltages, but this problem can apparently be solved by adding a hydrophobic monomer which is subsequently photopolymerized [90, 91].

4.2. *Phase separation*

In contrast to the emulsion method, phase separation techniques start with a one-phase mixture; phase separation is induced only after film formation, and it can be induced by cooling, solvent evaporation or polymerization [123]. The composites obtained are usually called PDLCs (for polymer dispersed liquid crystals).

4.2.1. *Temperature-induced phase separation*

4.2.1.1. *Description.*

It is possible to use this technique with thermoplastic polymers (e.g. poly(methyl methacrylate) [51], polyvinylpyrrolidone [51], polystyrene [124]) which are miscible with a liquid crystal at high temperature, but not at room temperature. The hot homogeneous solution of polymer and liquid crystal is coated on a substrate (sometimes with the help of a solvent which is evaporated at high temperature), and a second substrate is applied. During cooling, the components become incompatible and liquid crystal droplets are formed. Kinetics of phase separation seem to have a large influence on the structure of the composites [125, 126].

4.2.1.2. *Advantages and drawbacks.*

This technique is easily performed and it is possible to adjust droplet diameter by a careful control of cooling rate [21]. Additives, such as plasticisers [59], can be added to the polymer as long as they do not contaminate the liquid crystal [127]. On the other hand, the number of polymers with suitable solubility properties is limited. Even with such polymers, the fraction of liquid crystal which remains dissolved in the polymer binder has been reported to be as large as 40% [34], which dramatically modifies the polymer properties and uses a lot of liquid crystal. Finally, the structure of these composites is not stable with temperature.

4.2.2. *Solvent-induced phase separation*

4.2.2.1. *Description.*

A polymer and a liquid crystal can be used in this technique if they are not miscible and if they are both soluble in the same solvent. Such systems have been found, e.g. poly(methyl methacrylate) [24, 128] and polystyrene [129] in chloroform, and polycarbonate [130] in tetrahydrofuran. A solution of the polymer and the liquid crystal in the solvent is coated on a substrate. Phase separation occurs during evaporation of the solvent, and a second substrate can be applied.

4.2.2.2. *Advantages and drawbacks.*

Finding a suitable polymer and liquid crystal is easier than for the preceding method, but evaporation of the solvent is the critical step which determines the structure of the composite, and it is not straightforward to control the evaporation rate reproducibly. Moreover, during solvent evaporation, convection phenomena can create a spatial ordering and an orientation of the liquid crystal droplets [131].

4.2.3. *Polymerization-induced phase separation*

4.2.3.1. *Description.*

Polymerization-induced phase separation is by far the most studied method of preparation. It consists of introducing between two substrates a homogeneous mixture composed of the liquid crystal and a polymer precursor (monomers, oligomers, initiator, etc.); during polymerization the liquid crystal becomes insoluble in the medium. A very large choice of monomers is possible because the only restrictions are solubility of the monomers in the liquid crystal and refractive index matching. It is always possible to induce phase separation of the polymerized composite by adding crosslinking monomers. The variety of possible systems gives access to a wide range of properties [132]. Moreover, a wide range of liquid crystal concentration can be used because of the possibility of crosslinking the polymer. The liquid crystal content can be increased so that it constitutes the continuous phase: in this case the composite is sometimes called polymer-network liquid crystal [133–135]. If the polymer content is reduced even further (below 5%), then a gel-like material can be obtained [136]. Formation of the polymer binder can be induced by polymerization initiated photochemically, thermally or by electron beam [137, 138] or it can be induced by polycondensation.

4.2.3.2. *Free-radical polymerization.*

The large majority of radically polymerized systems are initiated by ultraviolet light. This versatile technique makes it possible to adjust kinetics of binder formation in several ways, so that very small droplet diameters can be obtained (10 nm [139]). The faster the polymerization, the smaller the liquid crystal droplets; this can be achieved with higher UV power [36] or with accelerators [140]. Crosslinker content also affects the structure of the composite [141]. Finally, polymerization temperature, which can be controlled, is very important [142] and a spinodal phase separation can be preferred in order to have a homogeneous size of droplets [143]. Polymerization and phase separation can be monitored by differential scanning calorimetry [144, 145] so that conversion and the quantity of phase separated liquid crystal can be determined. Photoinitiators have been shown to produce residues which contaminate the liquid crystal; this problem has been solved with copolymerizable photoinitiators [146]. Liquid crystal droplets can be deformed by applying a mechanical pressure during polymerization [105].

The main drawback of photochemically induced phase separation is its high sensitivity to experimental conditions, such as mixture composition, light power and polymerization temperature. This means that reproducible results can be obtained only by a careful control of these parameters.

4.2.3.3. *Polycondensation.*

Polycondensation is activated by heating a film of the precursors. The monomers most studied are epoxides [9, 63, 83] and isocyanates [147]. Kinetics of formation of the composites are much slower than in the free radical case, so that studies have been made by several techniques (e.g. light scattering [148, 149], calorimetry [147], viscosimetry [150]). Yet, the process is not easier to describe because formation of the composite is a function of polymerization rate, polymer solubility changes, diffusion of constituents and the isotropic to nematic transition temperature of the liquid crystal [151]. Density gradients between the liquid crystal and the prepolymer can also influence the structure of the composite because of sedimentation occurring before gelation [40].

Polymerization-induced phase separation is apparently very simple to use. It is true that a composite with reasonably good properties can easily be obtained, but improvement of these properties is a very complex step because nearly all electro-optical properties are interdependent.

5. Applications

Due to their versatile properties, liquid crystal–polymer composites are studied for a wide variety of applications. Among them, displays and smart windows are the most popular subjects in the literature.

5.1. *Displays*

Display applications can be divided into projection and direct view displays, because the configurations are different.

5.1.1. *Projection displays*

It is possible to display large images on a screen by projecting an image created on a small valve (figure 4). These diplays are schematically composed of a light source illuminating a liquid crystal–polymer composite valve: light rays that are not deviated pass through an objective and are projected onto a screen. On the other hand, divergent light rays cannot go through the objective, so that ON pixels are projected as white dots on the screen and OFF pixels correspond to black dots.

The main advantage of using a composite valve, rather than a conventional twisted-nematic light valve in projection displays, is that the on-state is one hundred per cent transparent because no polarizer is present [152]. Moreover, no surface treatment of electrodes is necessary so that fabrication is simplified.

The necessary degree of miniaturization and the fact that the threshold of the electro-optical curve is not sharp enough for multiplexing, imposes the use of active matrix addressing schemes. These active matrices are made from silicon and cannot deliver more than ten volts. Although cadmium selenide active matrices [153] can deliver up to a hundred volts and although it is possible to sharpen the threshold of the electro-optical curve [154], these solutions are not viable yet, so that a first specification for composites is to have a driving voltage lower than ten volts.

If video rate application is sought, then response times of the composite should be lower than 20 ms. Moreover, a contrast ratio of the order of 100 seems to be adequate. This contrast ratio has been shown to be a strong function of the angular dependence of the scattering curve [11, 152]. Finally, hysteresis should be as small as possible and these properties should be stable between at least 20 and 50°C. Several projection displays have been described in the literature [155, 156]. Although it is difficult to compare their properties because not all data are given, they show that liquid crystal–polymer composites are potentially interesting for projection displays.

5.1.2. *Direct view displays.*

In this case, the composite is used both as the valve and as the screen, so that it is vital to be able to make large area liquid crystal–polymer composite films. We describe three possible configurations to create a white and a black state, but there are other possibilities [61].

5.1.2.1. *Dichroic dyes.*

The composite is made from a liquid crystal containing a black mixture of dichroic dyes and it is placed on a coloured reflector (figure 5). In the off-state, orientation of the liquid crystal droplets is random, so that the composite scatters and absorbs incoming light: the screen is black. In the on-state, refractive indices are matched and the dyes are oriented and do not absorb, so that the composite is transparent: because of the reflector,

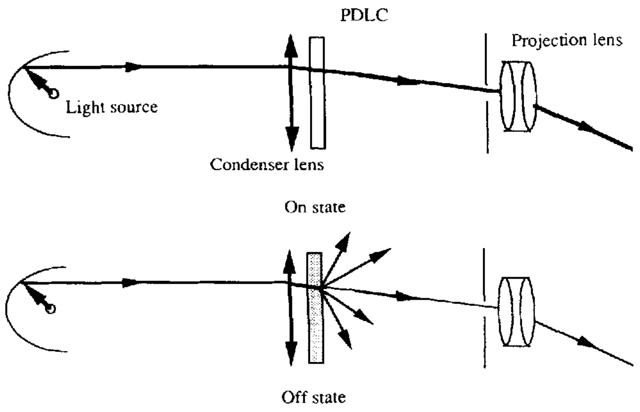

Figure 4. Schematic representation of a projection display.

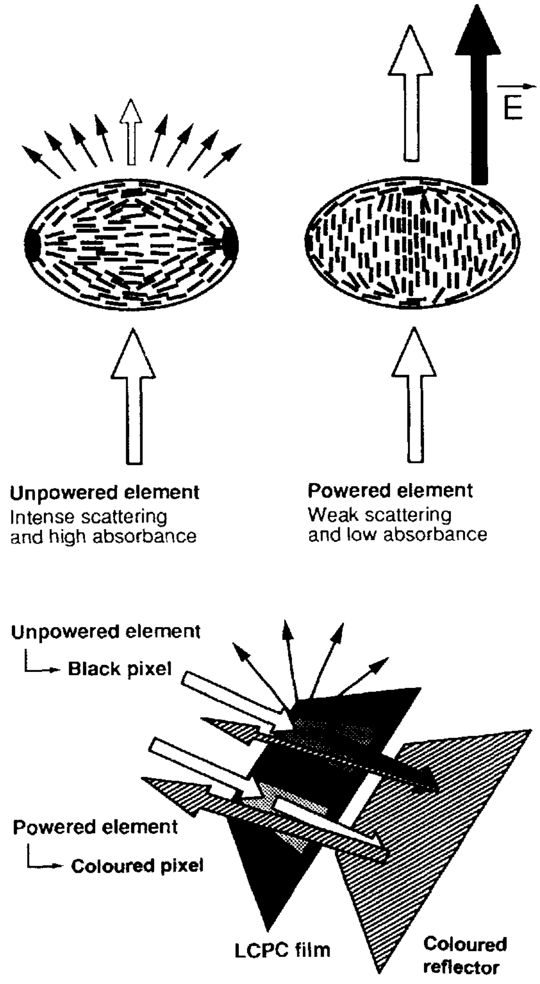

Figure 5. Schematic representation of a reflective display based on a liquid crystal mixture containing dichroic dyes.

the screen is luminous. There are several possibilities for making a coloured display on this principle [157, 158] and it is also possible to have a white scattering state rather than a white reflective one by introducing glass beads in the polymer [159].

Theoretically, this type of screen should have both high luminosity and high contrast, but in fact three problems are encountered. First, a good quality white state is difficult to obtain. Secondly, in order to have a high contrast, the dichroic dyes must be exclusively present in the liquid crystal droplets and not in the polymer binder, or else the contrast is the same as with isotropic dyes [160]. As a consequence, plasticising of the polymer by the liquid crystal itself must be as low as possible. Finally, photochemical processes cannot be used to make the composites because of the poor chemical and photochemical stability of dichroic dyes [161]. Azo dyes have been shown to have a poor UV stability [162] and anthraquinone dyes which are more stable have a lower dichroism [163, 164]. Thus, reflective displays containing dichroic dyes have been obtained by emulsion [165–168] or by polymerization induced phase separation of epoxides [169–171]. In particular, flexible coloured displays for touch panels have been fabricated. In addition to these restrictions on preparation, the low photochemical stability of dichroic dyes causes deterioration of the ageing properties of the composite [172].

5.1.2.2. *Light absorber.*

The screen is composed of a normal liquid crystal–polymer composite placed on a black absorber [110, 173–175]. In the off-state the screen is white because of back scattering, but in the on-state the composite is transparent so that the display is black. The main advantage of this configuration is its simplicity. Moreover, viewing angle is much better than for twisted nematic liquid crystal displays. The drawback of this display is its low luminosity which depends on the back scattering of the composite: only about 25% of the incoming light is back scattered.

5.1.2.3. *Crossed polarizers and back lighting.*

The composite is sandwiched between crossed polarizers, and a light source is placed behind the screen [176] (figure 6). In the on-state, the light from the source is blocked by the crossed polarizers: the screen is black. In the off-state, the light polarized by the first polarizer is depolarized by the birefringent droplets, so that the display is luminous. Although polarizers reduce the luminosity of the screen, this type of display is still interesting because it offers a better viewing angle than twisted nematic liquid crystal displays.

Thus, liquid crystal–polymer composites can be used in many configurations for display applications. The common advantage conferred by these materials is the possibility of making large area displays; the precise configuration to choose depends on the particular application intended.

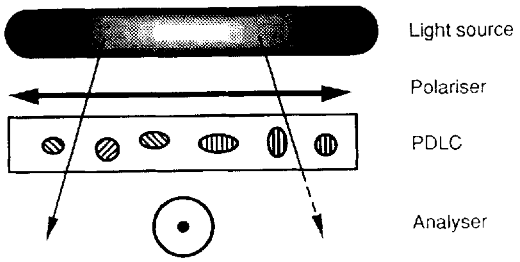

Figure 6. Schematic representation of a direct view display featured by crossed polarizers and back lighting.

5.2. *Smart windows*

5.2.1. *Normally scattering window*

Liquid crystal–polymer composites are good candidates to make privacy windows which can be electrically switched from opaque to transparent. Specifications are quite different from display applications: driving voltage and response times are no longer restricting properties. On the other hand, cost is a primary issue since large areas are envisaged. This means that plasticising of the polymer by the liquid crystal, which is the expensive component, has to be minimized. Moreover, large viewing angles are necessary.

5.2.2. *Normally transparent window*

For some applications, such as car windscreens, windows that are transparent in the off-state are required; several possibilities have been devised to meet this requirement. The first possibility involves using a polymer which induces a homeotropic anchoring and flattening the liquid crystal droplets [177]. In this case the stable configuration of the droplets is axial, with the axis perpendicular to the plane of the film. If the ordinary refractive index of the liquid crystal (n_o) is matched to the polymer index (n_p), then the film is transparent. Moreover, if the dielectric anisotropy of the liquid crystal is negative, the on-state is scattering.

Another way is to use liquid crystal mixtures whose dielectric anisotropy is negative at high frequency and positive at low frequency [178–180]. The composite is made by polymerization induced phase separation. During polymerization a low frequency electric field is applied, so that the liquid crystal is aligned perpendicular to the film; after polymerization and removal of the field, the liquid crystal remains oriented because of anchoring forces. Application of a high frequency electric field drives the film opaque.

Finally, photopolymerizing a few per cent of a difunctional monomer in a liquid crystal yields an anisotropic gel. If the substrates have been treated to induce planar anchoring, then the film is homogeneous and transparent. Application of an electric field makes the film opaque because only the liquid crystal which is not in contact with the polymer can rotate [181–183].

5.3. *Miscellaneous applications*

5.3.1. *Membranes*

Liquid crystal–polymer composites were studied as self-supported membranes for selective transport of gases and ions [184] before being considered for display applications [130]. The interest here lies in the high permeability jump occurring in the vicinity of the fusion temperature of the liquid crystal [185–187].

5.3.2. *Optical valves*

Optical valves are composed of a large layer of photoconducting material associated with a layer of liquid crystal–polymer composite. They can be operated in transmission [111, 188] or in reflection [189]. The two layers are sandwiched between two transparent electrodes, and a voltage is permanently applied. This device functions as a shutter for a red light beam. In the absence of an additional blue light beam, the photoconductor is resistive so that the voltage applied to the liquid crystal–polymer composite is low and the device is scattering. If a blue light is also present, then the photoconductor is conducting and the composite becomes transparent for all visible wavelengths.

5.3.3. *Bragg gratings*

In the case of phase separation induced by photochemical polymerization, it is possible to polymerize a mixture of monomer and liquid crystal through a mask [20] or with interference fringes [190–192]. The film obtained is composed of thin bands containing liquid crystal droplets of very different sizes; this structure diffracts light and the intensity of diffracted light can be electrically controlled.

5.3.4. *Memory effects*

Several types of liquid crystal–polymer composites present a memory effect [25, 28, 79, 132, 193, 194]. These composites are switched from a scattering state to a transparent state by applying a voltage, but if they are heated and cooled under an electric field, then they remain transparent after removal of the field. The scattering state can be recovered after heating and cooling without the electric field. This property can be used to make laser addressed displays [195].

5.3.5. *Infrared shutter*

Liquid crystals are birefringent in the visible spectrum, but also in the infrared [196], so that it is possible to make infrared shutters [197]. Of course, some wavelengths have to be excluded because of absorption by the liquid crystal and polymer.

5.3.6. *Gas flow and pressure sensors*

Liquid crystal–polymer composite films with one side exposed have been studied as gas flow sensors for aerodynamic testing [198, 199]. Orientation of the liquid crystal droplets at the surface of the film is a function of shear stress and can be monitored by optical measurements. If an indium tin oxide coated polyvinylidene fluoride film is put on a composite film, and if the liquid crystal contained in the composite is a ferroelectric, then a pressure sensor is obtained [200].

5.3.7. *Solar protection*

Liquid crystal–polymer composites seem to be good candidates for solar protection, because they do not absorb light and so are not heated up [140, 201]. In this application, back scattering has to be optimized and the composite must be protected from ultraviolet light.

5.3.8. *Non-absorbing polarizers*

If a liquid crystal–polymer composite film is stretched, droplets are elongated and the liquid crystal inside the droplets is aligned in the direction of stretching. This composite is a good polarizer because only the polarization perpendicular to the stretching direction is transmitted [202]. The other polarization is not absorbed but scattered, so that the film is not heated even under large light power.

This list of applications for liquid crystal–polymer composites is certainly not exhaustive. It is even less so if we consider the following composites that are related to the conventional nematic liquid crystal–polymer composites.

6. Related composites

6.1. *Composite with a polymer liquid crystal binder*

We have seen (§ 3.3.1.4) that it is possible to reduce haze at large viewing angles by using a polymer liquid crystal binder with both refractive indices matched to those of the low molecular weight liquid crystal ($n_{op} = n_0$ and $n_{ep} = n_e$). This concept has been checked with epoxides [203–206] or polysiloxanes [207] bearing mesogenic side chains. By choosing side chains chemically similar to the low molecular weight liquid crystal, it is possible to match the indices, but phase separation is not complete.

6.2. *Composite with very little polymer*

We have already seen (§5.2.2) that anisotropic gels [208, 209] obtained by photopolymerization of a few per cent of monomer in a liquid crystal can give transparent composites in the off-state. In fact, it has been shown that the polymer is oriented by the liquid crystal during polymerization [210–212]. That is why the effect of an even smaller proportion of polymer (0·1%) in twisted and supertwisted nematic liquid crystal displays [213–216] or in smectic displays [217] has been studied. This very small quantity of polymer does not destroy ordering of the mesogens, but it provides a good anchoring which could be an alternative to usual surface treatments.

6.3. *Composite with a cholesteric liquid crystal*

Until now, cholesteric liquid crystal–polymer composites have been less studied than their nematic homologues, but they seem to offer even more possibilities. Indeed, depending on electric field and anchoring conditions, cholesteric composites can have a scattering focal conic structure, a transparent homeotropic texture or even a coloured Granjean texture which reflects one circular polarization at a wavelength associated with the pitch of the liquid crystal. Consequently, several electro-optical effects have been demonstrated. It is possible to switch from a scattering or a transparent state to a coloured one by increasing the voltage [183, 218–221]. In the case of cholesteric gels, it is also possible to obtain bistable states which are scattering, coloured or transparent [222–225]. Switching between these states is achieved by applying well defined voltage pulses. Based on this principle of operation, several types of display have been demonstrated. For instance, advertizing displays, changeable message signs and pagers are commercially available.

Moreover, there is a richer number of configurations of cholesteric liquid crystals confined in droplets than in the case of nematics [226–230], and the possibility of using a flexoelectric-based electro-optical effect has been demonstrated. [231].

6.4. *Composite with a smectic liquid crystal*

The electroclinic effect [232, 233] and the unwinding of the ferroelectric or antiferroelectric helix [231, 234, 235] have been used to induce birefringence variation in liquid crystal–polymer composites. A surface treatment or application of shearing during polymerization is necessary to orient the liquid crystal droplets. The advantage of these materials is their short response time (about 0·1 ms).

6.5. *Composite with an inorganic binder*

Composites with an inorganic binder have recently been shown to present the same electro-optical effect as conventional composites. They are prepared by sol–gel processes [236–238] or by filling a porous alumina film with liquid crystal [239]. Refractive index matching, which can be difficult, seems to be achievable by doping of the binder with high index titanium oxides [240].

Another way to use ceramics is to disperse a fine silica powder (50 nm) in the liquid crystal [241]. This powder is too fine to scatter light by itself, but it creates defects in the liquid crystal which are scattering. Anchoring is so strong that it is necessary to apply a voltage and heat (laser) to switch the liquid crystal.

6.6. *Miscellaneous composites*

6.6.1. *Photosensitive dopant*

Dopants consisting of azo compounds which are in a *trans*- or *cis*-configuration, depending on the wavelength of incoming light, have been incorporated in liquid crystal–polymer composites [242, 243]. The dopant in the *cis*-configuration destabilizes the mesogen, so that the composite can be maintained at a temperature between the clarification temperatures of the liquid crystal doped, respectively, with dopant in the *cis*- and in the *trans*-configuration. At this temperature, the liquid crystal is nematic or isotropic, depending on the wavelength of the incoming light, and thus the composite is scattering or transparent.

6.6.2. *Effect of a high power laser*

Effects of a high power laser on liquid crystal–polymer composites have been studied by several groups [57, 244–249]. In particular, G. Cipparrone *et al.* [250] have measured the transmission of a laser beam across a composite containing a dichroic dye absorbing at the wavelength of the laser: if the power of the laser is raised above a threshold, the liquid crystal becomes isotropic and scattering is considerably reduced.

6.6.3. *Thermo-optical effect*

We have already mentioned the use of liquid crystals whose dielectric anisotropy is positive or negative as a function of frequency (§3.3.3.3, 3.3.4.3, 5.1.1 and 5.2.2). The frequency f_c at which $\Delta\varepsilon = 0$ is itself a function of temperature [251], so that if the composite is addressed at a frequency close to f_c, it is transparent or scattering depending on slight temperature variations [252].

7. Conclusion

Much progress has been achieved during these last ten years in the understanding of LCPCs. Their improved electro-optical properties allow the expectation of practical realizations in the next few years. Until now, photopolymerization-induced phase separation has been the most studied method to prepare LCPCs, and nematic mixtures leading to driving voltages lower than

7*V* are now commercially available. The potential interest of nematic LCPCs in projection and direct view displays has been demonstrated by several groups, but for LCPCs to replace existing technologies, a higher contrast together with a low driving voltage have to be attained. A real breakthrough will be achieved when an LCPC with an adapted scattering profile becomes available. In fact, this is a serious issue, since no theoretical model taking into account the size, the shape and the density of the liquid crystal domains is at our disposal as a guide for LCPC preparation. In addition, little is known concerning the required nature of the interface between the liquid crystal and the polymer itself to obtain LCPCs of practical interest.

Large-area architectural light shutters obtained via an emulsion method were commercially available some years ago. But the off-axis haze and the photochemical stability of the materials remain problems for nematic LCPCs; these have to be solved to allow their future come-back in the smart window market. Finally, cholesteric gels seem very promising for direct view displays provided that a lowering of their driving voltage is achieved.

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