UV Cured Flexible Cholesteric Liquid Crystal Displays

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Abstract

Flexible Cholesteric liquid crystal displays have been rapidly maturing into a strong contender in the flexible display market. Encapsulation of the Cholesteric liquid crystal permits the use of flexible plastic substrates and roll-to-roll production. Recent advances include ultra-thin displays, laser-cut segmented displays of variable geometry, and smart card applications. Exciting technologies such as UV curing, and simultaneous laser-edge sealing/singulation enable high volume production, excellent quality control and non-traditional display geometries and formats.

Keywords: UV Cure, Flexible Displays, Roll-to-Roll, Plastic Substrates, Cholesteric Liquid Crystals, Bistable

Introduction

Nearly every commercial liquid crystal display (LCD) manufactured and sold today is made from a sandwich of liquid crystal (LC) between two precisely spaced glass substrates coated with a transparent conductor. A few challenges remain for LCDs in the consumer market; namely, flexibility, durability, weight and power consumption. The first three issues can be addressed by using thin plastic substrates whereas the last issue can be solved using a bistable display, i.e. a display that is stable (with no power applied) in two states such as reflective or transmissive. Unfortunately, a problem arises when sandwiching liquid crystal between two flexible substrates: the fluid will flow! When the cell is flexed or pressed upon, the gap between the substrates becomes smaller displacing the LC and inducing flow.

Cholesteric liquid crystal displays (ChLCDs) are bistable LCDs that have low power consumption and exhibit high reflectance under ambient lighting[1]. As such, they are suitable for numerous applications from small handheld devices to large area signage. Indeed, the most recent consumer products utilize glass ChLCDs for portable flash/hard-drive storage devices, Figure 1. ChLCDs are desirable for flash-drive applications because the bistable display is only used to display current memory capacity from the last USB-powered update. The display requires no power to retain the image so it remains in the reflective state until plugged back into the USB and updated. As with most commercial LCDs, ChLCDs are typically made on glass substrates. The need and desire to use plastic substrates in place of the typical glass substrates has been there since the beginning of volume LCD manufacturing. However, this change requires significant effort. ChLCDs can easily utilize plastic substrates [2], they do not require polarizers (there is no condition for non-birefringent substrates),[3] and they do not require precise anchoring alignment control as do twisted nematic/super-twisted nematic LCDs. The move to plastic substrates allows one to make significantly thinner LCDs, more rugged, light weight, and ultimately flexible.[4]

Figure 1. Glass ChLCDs in USB flash drive (right) and portable hard drive (left) applications.

We have found that in order to create robust flexible ChLCDs using plastic substrates, one must encapsulate the liquid crystal materials.[5] Encapsulation prevents the LC from flowing and also adds mechanical strength to the plastic display. The two most common methods to encapsulate liquid crystals are by phase separation (which is used with Polymer Dispersed Liquid Crystals (PDLC) systems)[6] and emulsification (or microencapsulation) of the liquid crystal in a binder.[6,7] Both methods have their advantages and disadvantages: however, this paper will focus on phase separation.

Encapsulation via Polymerization Induced Phase Separation

During polymerization induced phase separation (PIPS), droplets of liquid crystal are excluded from the bulk via phase separation as the polymeric chains grow in molecular weight.[7] The process of phase separation is a dynamic process. Initially, it is important that the prepolymer composition containing monomer, photoinitiator and liquid crystal be solubilized to the homogeneous single phase so the process starts with uniformity.[7] The mixture is laminated to a precise spacing between plastic substrates and then exposed to a low irradiance UV-A lamp. During photo-polymerization, the LC is excluded from the bulk as the polymer increases in molecular weight. Liquid crystal droplets begin to coalesce during the phase separation process. The polymer solidifies into encapsulating walls that completely entomb the liquid crystal. The speed at which the LC-polymer dispersion polymerizes is crucial to controlling the uniformity and size of droplet formation. One advantage of the process is that there are a number of variables that can be adjusted to control the speed of photo-polymerization. One can adjust the number of functional groups per polymerizable molecule (crosslinking density), the reactivity of the functional groups, the concentration of the photoinitiator, the monomer to liquid crystal volume fraction, the monomer size (~viscosity), the cell spacing, and the irradiance of the UV light.
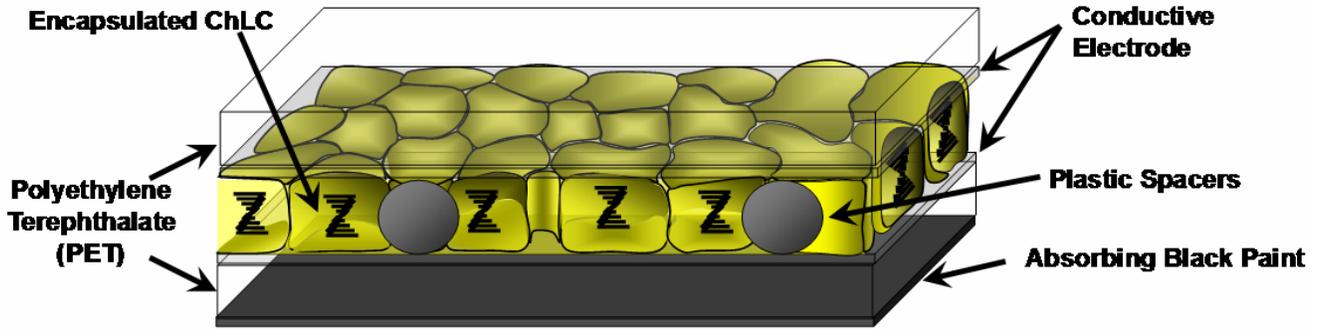
Figure 2. *Cross-section of a PIPS-encapsulated Cholesteric Liquid Crystal Display.*

The cross-section of a PIPS-encapsulated Cholesteric liquid crystal display device is seen in Figure 2. The droplets that are created from this process differ significantly in morphology when compared to traditional polymer dispersed liquid crystal (PDLC) displays. In most PDLC-type displays, the droplets are usually spherical, less than 1 micron in diameter, and are numerous in number throughout the thickness of the cell.[6] In our PIPS process, the droplets are more 'pancake' in shape, on the order of ~10µm (or more) in diameter and 4µm in height. It is important to note that there is typically only one droplet throughout the thickness of the cell, as can be seen in the SEM micrograph of the PIPS wall structure in Figure 3b (the top substrate and liquid crystal were removed before imaging). Having only one droplet throughout the thickness of the cell reduces light scattering from the LC-polymer interface. Notice the 4µm spherical spacers embedded in the polymer wall in Figure 3a compared to the droplet diameter.
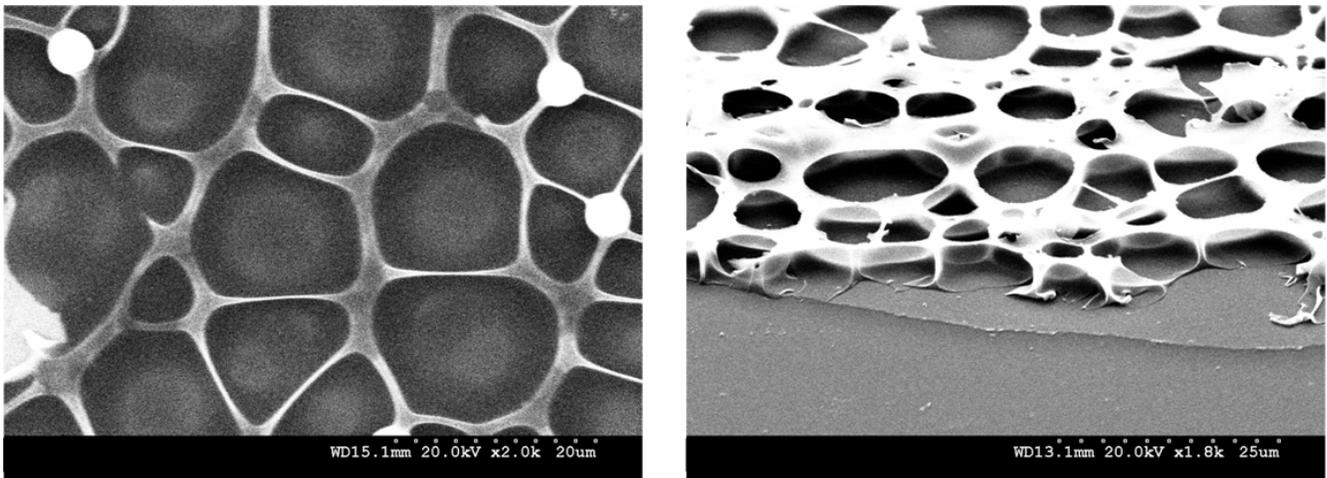
**Figure 3. a.)** *The polymer encapsulating walls are evident in the SEM micrograph at left. Notice the 4µm spherical spacers embedded in the droplet wall.* **b.)** *The pancake-shape of the droplet is evident in the SEM micrograph at right.*

Cholesteric Liquid Crystals

A nematic thermotropic liquid crystal is a melt composed of molecules that are typically elongated and freely exhibit at minimum some thermodynamically stable mesophase where the molecules are locally ordered (oriented) within the fluid.[8] The Cholesteric liquid crystal (ChLC) is

created when a chiral dopant of a particular handedness is added to the nematic liquid crystal in sufficient concentrations such that it twists the nematic molecules 180° within a distance of hundreds of nanometers.[8] The ChLC can be electrically switched to either one of two stable textures; planar (reflective) or focal conic (transmissive). Light is selectively reflected from the planar texture of the Cholesteric liquid crystal. Due to the selective reflection, red, green and blue reflecting ChLC layers (as well as IR reflecting layers) can be stacked on top of one another without optically interfering. The director (long axis of the molecule) of the planar texture is uniformly parallel to the plane of the substrates across the cell but has a helical twist perpendicular to the plane of the substrates. It is the helical twist of the uniform planar texture that Bragg reflects light of the same helicity (handedness) in a selected wavelength band.[8] The maximum theoretical reflection achievable with one Cholesteric layer is 50% of the light for a LC with a chiral dopant of one handedness, i.e., left or right handed. Interestingly, two layers of the same color but of opposite handedness can be stacked on top of each other to nearly double the reflectivity.
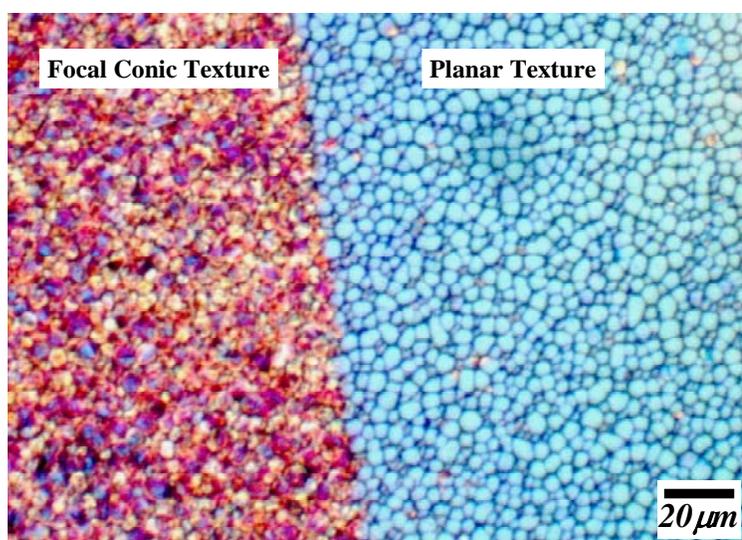

Figure 4. *A polarizing micrograph between crossed polars of a PIPS-encapsulated ChLC. Notice the uniform planar texture (right) and non-uniform focal conic texture (left).*

The focal conic texture contains the so-called "focal conic" defects that perturb the orientation of the liquid crystalline helices. In the typical focal conic texture, the defect density is high thus the helical domain size becomes small and randomized in orientation such that it is just forward scattering and does not reflect incident light. When the LC is firmly anchored to the surface and the focal conic defect structures are created, they are topologically stable and cannot be removed unless by some external force such as an electric field. Thus, the focal conic texture remains stable and forward scatters light of all wavelengths into an absorbing (usually black) background. The bistable textures of the ChLC can be electronically switched between each other at rapid rates (on the order of milliseconds).[8] Gray scale is also available within a single pixel through various switching schemes in order to adjust the density of reflective planar domains that are oriented perpendicular to the substrates to the slightly scattering focal conic domains that are randomly oriented. In Figure 4, we can see a micrograph of a PIPS encapsulated ChLC display using non-birefringent polycarbonate substrates. The image was taken using the transmission mode of a polarizing microscope between crossed polars. The focal conic texture on the left in Figure 4 appears multicolored and non-uniform due to the birefringence effects of the focal conic

texture.   The planar texture on the right appears uniform due to the planar alignment of the liquid crystal. The polymer walls appear as black lines because they are not birefringent –the light is extinguished as the sample is illuminated between crossed polars.
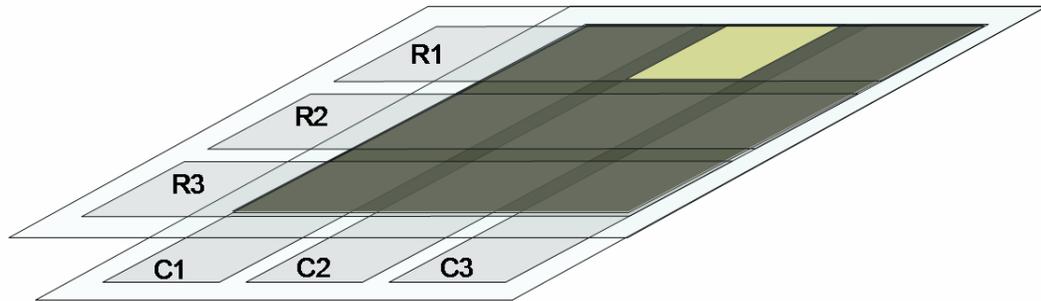
Figure 5. *For passive addressing, a pixel is written by addressing rows and columns.*
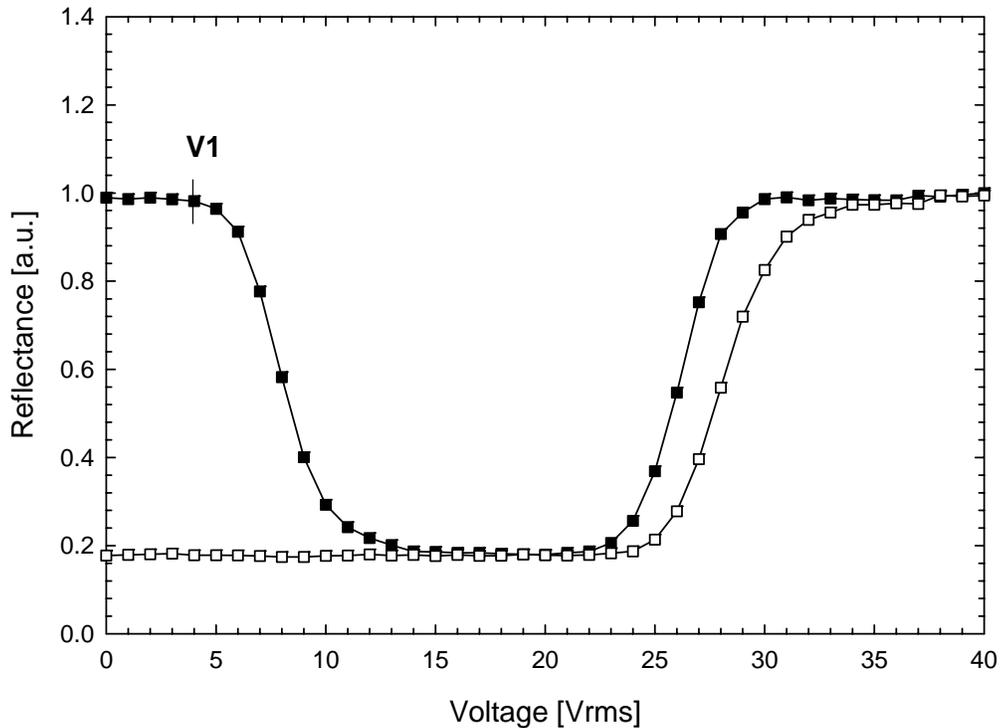
Figure 6. *Curves for a 4um thick cell showing the planar (solid square) and focal conic (white square) electro-optic response. Notice that the threshold voltage to switch from the planar state to the focal conic is V1~4V.*

Pixelated displays are typically driven with an active matrix of thin film transistors (TFTs) at each pixel. The active matrix TFT is a very expensive display architecture. An alternative is to passively address each pixel by the intersection of conductive rows and columns (Figure 5). Bistable Cholesteric liquid crystal displays have interesting electronic drive features that other bistable reflective technologies do not. They can be easily passively driven. The threshold voltage, V1, as seen in figure 6, is very important when addressing a matrix of many pixels within a display. A threshold voltage is essential for multiplexing (passively addressing) a row/column matrix without the need for an expensive active matrix (transistor at each pixel) backplane. In Figure 5, a pixel is written by applying a column

voltage to the bottom plane conductors (C1, C2, C3) and a much higher addressing voltage across a single row (R1) conductor. If the column voltage is above the threshold voltage ($V_c > V1 = 4V$), pixels other than the intended one to be addressed will begin to change to the focal conic state. Bistability with a voltage threshold allows high-resolution ChLCDs to be manufactured on plastic substrates and addressed using low-cost passive matrix electronics.

UV Photopolymerization, Functionality and Droplet Morphology

Electro-optic performance parameters such as reflectivity, switching voltage, optical contrast ratio, and response time in a PDLC device can be controlled through prepolymer formulation. These parameters are related to the shape, volume, distribution and size of the liquid crystal droplets and surface anchoring and director orientation of liquid crystal in the droplets. In case of photopolymerization induced phase separation of polymer in liquid crystal, these parameters are related to monomer concentration, monomer functionality, photoinitiator concentration, light wavelength and intensity, and polymerization temperature. At fixed polymerization conditions such as temperature and light source (of course, assuming constant pressure and volume), the polymerizable composition and the function of the polymerizable additives play major roles in the electro-optical properties of the PDLC device. We control photopolymerization chemically through the fraction of the crosslinker in the monomer/LC mixture, i.e., controlling the rate of polymerization through the average number of polymerizable groups in the pre-polymer. The molar functionality (F_{mol}),

$$F_{mol} = \sum_{x=1}^{n} f_x * M_x * (1mol_x / MW_x) \bigg/ \sum_{x=1}^{n} M_x * (1mol_x / MW_x),$$

approximates the rate of polymerization by calculating the average number of polymerizable groups per molecule. f_x is the number of polymerizable groups on monomer x. M_x is the mass fraction of monomer x in the total polymerizable mixture and MW_x is the molecular weight of monomer x. A higher functionality yields a faster polymerization rate and higher crosslinking density, which results in a smaller droplet size because of less diffusion time before the polymer solidification.

Photopolymerizable dispersions are prepared by mixing the desired composition of monomer, crosslinker, and photoinitiator in a vial with a non-polymerizable liquid crystal at room temperature.[5] The prepolymer (PP) to LC ratio is typically around 20:80, by weight %. Once this is vortex-mixed, 4µm plastic spherical spacers are added to the homogeneous single-phase mixture to preserve substrate spacing before polymerization. The prepolymer and liquid crystal mixture is laminated between two Polyethylene Terephthalate (PET) substrates coated with conductive electrodes to the cell gap spacing determined by the spacer size. The material is polymerized under a low irradiance UV-A lamp. After polymerization, the cell consists of well-encapsulated Cholesteric liquid crystal droplets that can be electrically switched to reflective, transmissive or gray-scale (partially reflective) states.
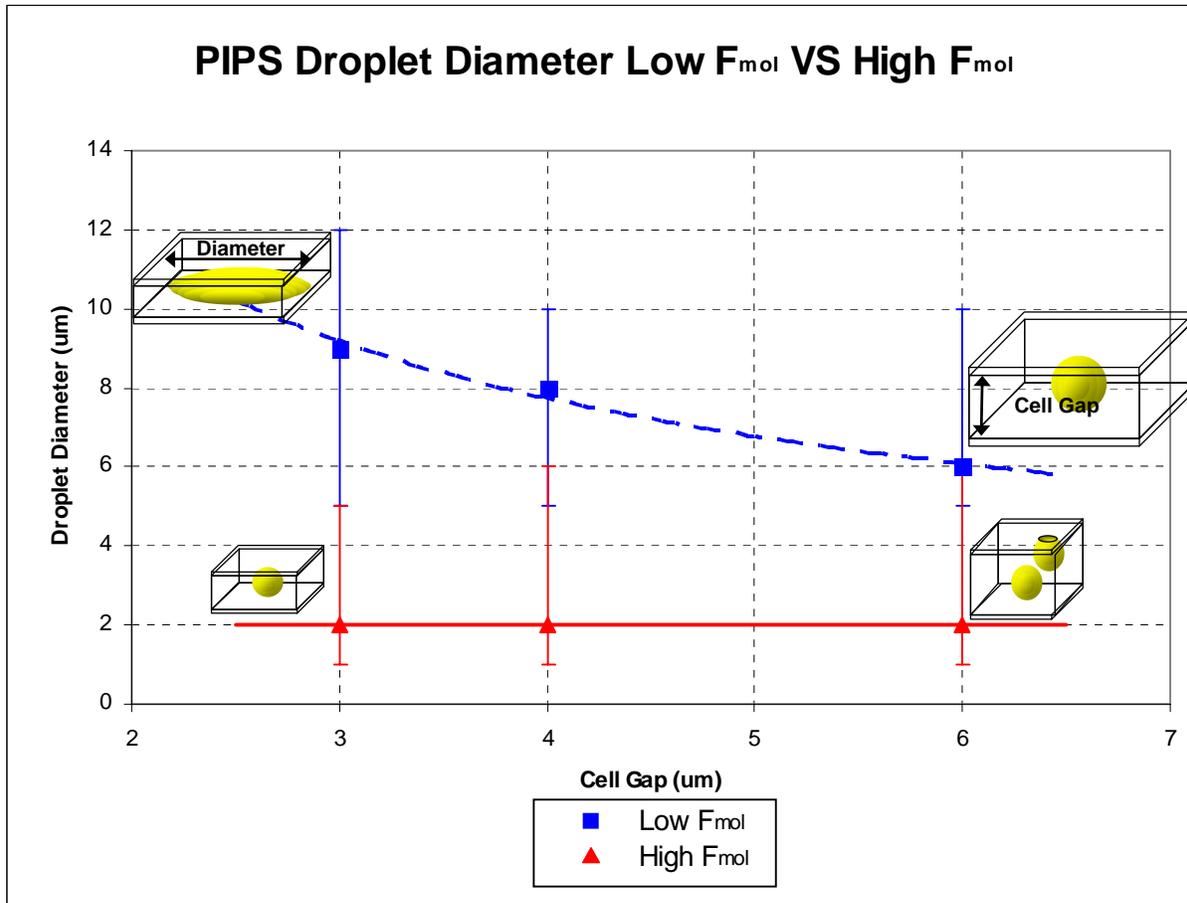
Figure 7. *Droplet diameter (micrometers) and shape for two functionalities (one low, the other high) within a 3, 4, or 6μm thick cell (cell gap). Notice that the higher functionality cells have spherical droplets that are smaller than the cell gap in diameter whereas the lower functionality cells have pancaked droplets at smaller cell gaps.*

The average droplet diameter was measured by optical microscope for each cured PIPS sample (of a particular functionality and cell gap) and plotted in Figure 7. The error bars show the largest and smallest droplets in any given field of view. Figure 7 shows that for a sufficiently high functionality, the average droplet diameter is smaller than the cell gap resulting in spherical droplets (inset lower left). Notice as the cell gap increases for the high functionality mixture, the droplets don't change their size, they just become more numerous through out the thickness (inset lower right). For lower functionalities, the droplets are pancaked at smaller cell gaps (inset top left) and spherical (inset top right) for sufficiently large cell gaps. More or less, the functionality controls the droplet volume and the cell gap determines the droplet shape. In Figure 8, we can see the reflectivity spectra (measured by an integrating sphere) as a function of cell gap for the low functionality mixture –notice that pancaked shapes enhance reflectivity. In Figure 9 we see a plot of the peak reflectivity for a given functionality as a function of cell spacing. Notice that the low (~18%) reflectivity for the high functionality mixture in Figure 9 is almost independent of cell gap.
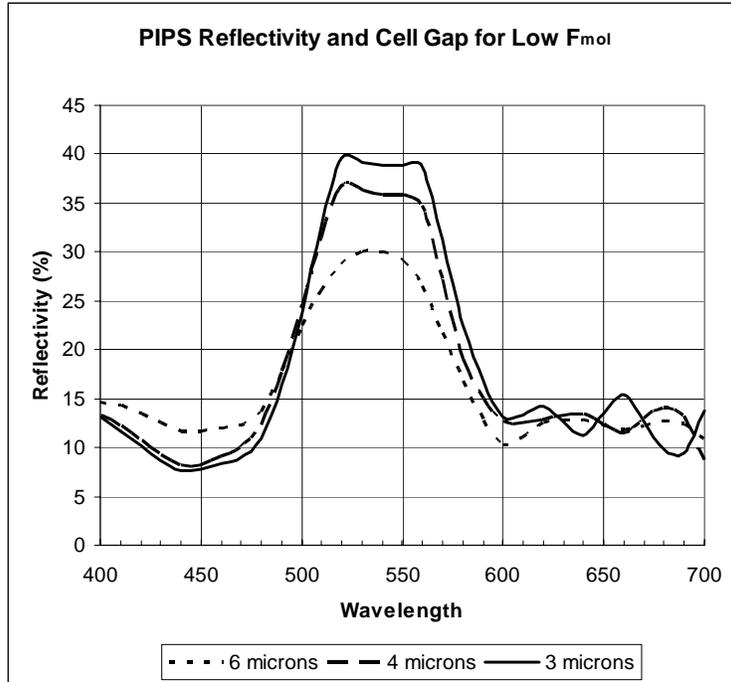
Figure 8. *Reflectivity spectra for a given cell gap for a Low F_{mol} PIPS Cell.*
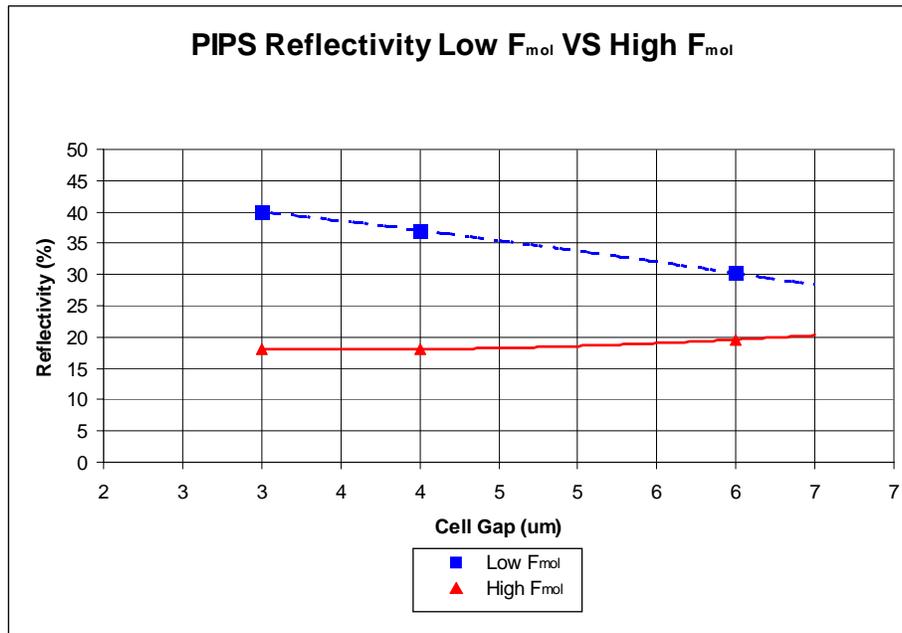
Figure 9. *Reflectivity of the planar texture as a function of cell gap and functionality.*

The ratio of pre-polymer to liquid crystal in the polymerizable mixture can be used to control the droplet size and polymer wall thickness. In Figure 10, the wt. % concentration of the pre-polymer in the liquid crystal is varied for two given functionalities. The lower functionality material forms larger droplets compared to the higher functionality material. Droplet sizes decrease as the concentration of

polymer in the system is increased. By increasing the amount of polymer in the system, we can control the thickness of the droplet walls (inset, Figure 10) and modify mechanical properties of the cell.
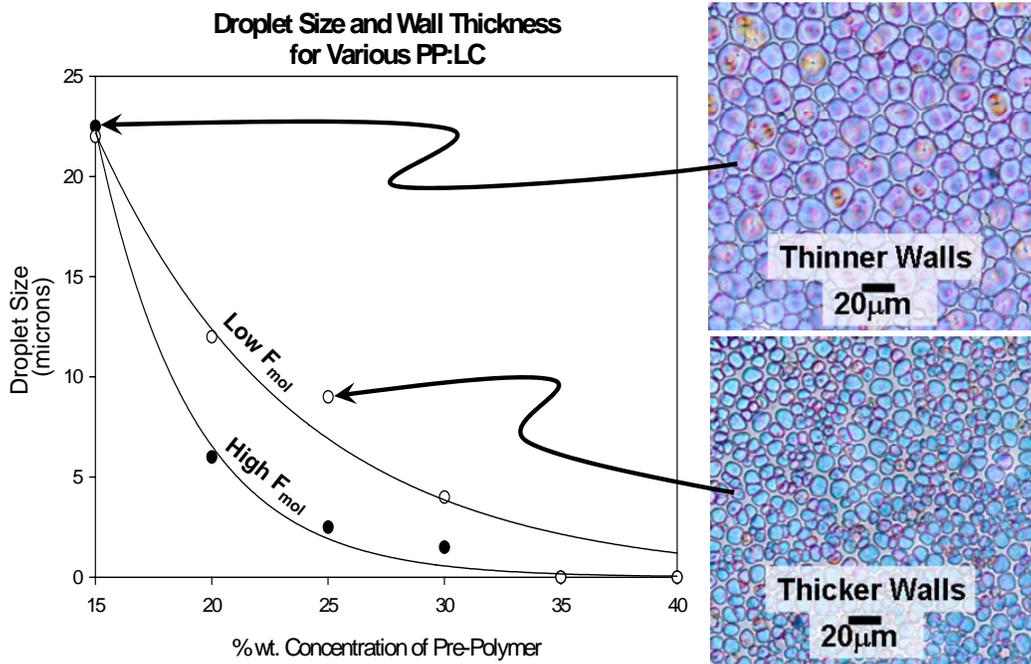
Figure 10. *Droplet size (diameter) of encapsulated ChLC as a function of pre-polymer concentration. Notice that the material with thicker walls has a higher proportion of pre-polymer (micrographs, inset right).*

Laser Singulation and the Roll-to-Roll Process

One motivation to use plastic to make LCDs is to manufacture the displays in a continuous roll-to-roll process. For roll-to-roll processing to be effective, the liquid crystal material should be filled between two spaced substrates and prevented from flowing by an encapsulation process. They should then be singulated from the sheet into the desired shape.

The polymer walls created during the encapsulation process are not very strong since they are typically sub-micron in width. Due to the thin polymer walls and the fact that there is very little solid material to hold the substrates together, the two bounding substrates are susceptible to delamination at the edges of the LCD. In addition, the edges should be sealed to prevent environmental contamination such as from humidity.

We have developed a method to simultaneously laser singulate and edge seal the display. The primary benefits to using laser singulation in the roll-to-roll process are efficiency and speed. The process uses a CO_2 laser that cuts the substrate and laser welds the edges together. The laser welds add sufficient strength to the edges of the display such that it does not delaminate and can be considered sealed. The laser is fed the cut parameters from a CAD file and thus retooling to cut a different shape is not necessary.

Displays of Unusual Shape

PIPS Displays built using the laser singulation process can be cut to unconventional shapes and formats. Figure 11 shows a laser singulated "KENT" display and a "spiral" display where the letters are active pixels. The "spiral" display was made from patterned ITO on 2 mil thick PET and the unconventional shape allows for a helical spiral when hanging (inset, Figure 11).


Figure 11. *Laser edge sealed ITO-based "KENT" display (left) and "spiral" display (right). These demonstrate thru-holes and non-conventional shapes that are not possible with glass displays.*

Full Color Pixelated Multi-layer Displays

As mentioned previously, Cholesteric liquid crystal displays achieve color by stacking selectively reflective layers, e.g., red, green, and blue (RGB) layers can be stacked for full color. Figure 12 shows an ultra-thin (~70μm) 24×20 pixel 3-layer (RGB) display using PEDOT-based conductive polymer for the electrodes. The display was made with ½ mil thick PET substrates that have inkjet printed conductive polymer electrodes.[12] The display was laser singulated from a panel of 4 displays. In figure 13, we can see a 3 layer (RGB) display that has been laser cut diagonally across the active portion of the display. Notice it can still be electrically switched. It was passively addressed to show a United States flag pattern after the cut, Figure 13.
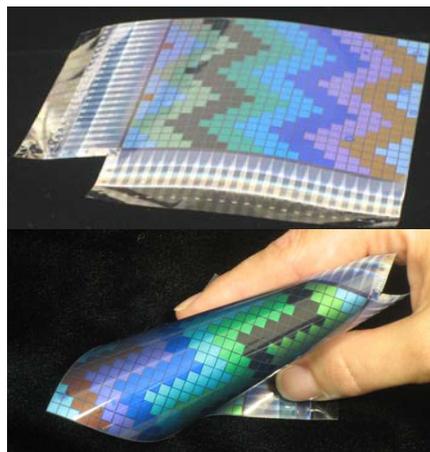
Figure 12. *Above: A laser edge sealed 24×20 pixel 3 layer ultra-thin (~70μm) display. The display edge at the left corner is within 200μm of the active area. Below: the display being flexed.*
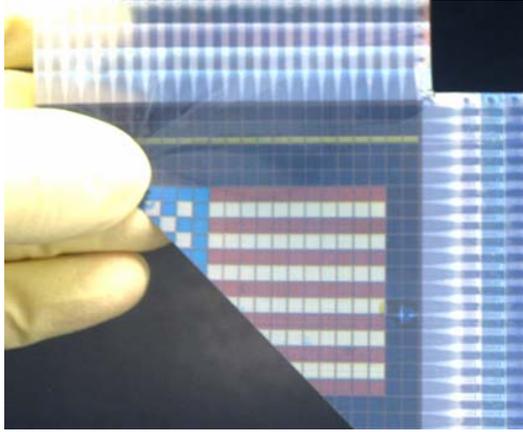
Figure 13. *A multi-layer 24×20 pixilated display that was cut diagonally through the active area demonstrating multiplexing.*

Conclusion

Flexible Cholesteric liquid crystal displays have been rapidly maturing into a strong contender in the future flexible display market. Encapsulation of the ChLC via photo-polymerization permits the control of droplet morphology and the ability to tune the electro-optic response. Encapsulation also permits the use of flexible plastic substrates and roll-to-roll production. The synergy of ultra-thin, variable geometry displays that can be retooled for different formats on the same manufacturing line hints towards a disruptive display technology in the near future.

Acknowledgements

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