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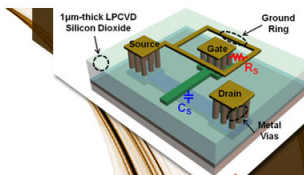
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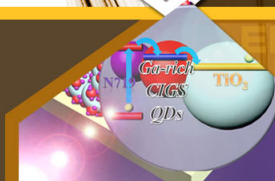
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Homeotropically aligning phase separated columnar structures for fabrication of flexible electrooptical devices

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A method of achieving homeotropic alignment of liquid crystals (LCs) by ultraviolet light induced phase separation of LC from its mixture with photo-curable pre-polymer is reported. Vertical polymer columns and micro-fibrils developed during the phase separation promote homeotropic alignment of the LC director (i.e., alignment perpendicular to the LC-substrate interface), suitable for devices based on LCs possessing negative dielectric anisotropy. These vertical structures extend between two substrates and permit the fabrication of highly flexible electro-optical devices with high contrast coupled with fast response times. This simple single-step technique eliminates the need for the traditional polymer alignment layer pre-deposited on substrates. © 2011 American Institute of Physics. [doi:10.1063/1.3663966]

In recent years, the need of and interest in developing flexible liquid crystal displays (LCDs) for applications, such as smart cards, cell phones, personal digital assistants, e-books, head mounted displays, etc., has grown exponentially because of their light weight, flexibility, and the potential for inexpensive roll-to-roll processing. Different electro-optical modes, such as twisted nematic, super-twisted nematic, and in-plane switching, have been employed to fabricate prototype LCDs using flexible substrates.¹⁻³ None of the conventional fabrication methods can ensure uniform separation between two flexible substrates, primarily due to non-uniformity in the cell thickness caused by the movement of spacers and mechanical deformations, thus making them unsuitable for fabrication of flexible devices. Recently, several methods that involve various forms of liquid crystal (LC)/polymer mixtures have been developed for the fabrication of LCDs using flexible substrates. These include polymer dispersed liquid crystals (PDLCs), phase separated composite films,⁴ formation of polymer walls assisted by electric field⁵ or induced by surfaces,⁶ pinning of spacers by a polymer film during phase separation,⁷ and photolithographically patterned polymer structures on alignment layers.⁸⁻¹⁰ Since these approaches do not provide bonding between two flexible substrates, the cell gap of LCDs fabricated using these techniques do not maintain uniform thickness when flexed, and therefore, most of these approaches have not been implemented in mass production of flexible displays. The PDLC-based LCDs on plastic substrates are more flexible than the other types of displays, but they operate in the light scattering mode, requiring high operating voltages, and have low multiplexibility, making them less useful for most display applications.

In this letter, we report a simple one-step method of preparing LC-based flexible electro-optic devices by simultaneous phase separation of LC and polymer, and director alignment via the formation of vertical polymer columns

upon exposure to ultraviolet (UV) light. These polymer columns, with similar but thinner polymer micro-fibrils in the inter-columnar space, not only bond the two substrates together to provide uniform cell thickness and flexibility, but also align LCs perpendicular to the substrates (i.e., homeotropically). Using LCs with negative dielectric anisotropy, we have fabricated LCDs with high flexibility, high contrast, low operating voltage, fast switching speeds, and, most importantly, without the use of any alignment materials and surface treatments. This approach of forming columnar polymer structure and achieving homeotropic alignment of LC offers a number of advantages over the conventional methods. First, a uniform alignment of LC can be achieved without depositing any alignment material on the substrates. This avoids a number of problems related to the deposition, thermal crosslinking, and the cumbersome rubbing technique required for the specific polymer materials used. Second, since the devices made with this method are based on vertical director alignment, the OFF state is completely dark and the brightness of the ON state can be adjusted by selecting LCs with higher birefringence and tuning the cell thickness to obtain very high contrast. Third, the microscopic polymer columns bond to the two substrates making the flexible displays mechanically robust and insensitive to deformations and external pressure.

In the simplest form, formation of the columnar structure involves the fabrication of a LC cell with two glass substrates coated with transparent indium tin oxide (ITO) electrodes. The substrates are held together forming a thin (typically 3–5 μm) cavity determined by the diameter of the glass fiber spacers. A mixture of photocurable prepolymer NOA65 (or NOA68, NOA77, Norland Products Inc.) and chiral-doped nematic LC material MJ 991213 (MJ) with negative dielectric anisotropy (Merck Korea, Ltd.) mixed in the ratio 1:1 is filled into the cell by capillary action at 95 °C. The LC cell is then exposed to $\sim 1.3 \text{ mW/cm}^2$ UV light at 360 nm at the same temperature. Illumination by UV light initiates

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polymerization along with crosslinking of the growing polymer chains, which leads to phase separation of the LC from the polymer matrix. Phase separation depends, among other parameters, upon the interplay between the surface interactions and the inter-diffusion of LC and prepolymer at the molecular level.¹¹ The pre-polymer preferentially wets (and bonds to) the ITO surface and polymer micro-droplets begin to form at the interface during photo-polymerization. These micro-droplets act as nucleation sites where additional polymer mass is deposited forming vertical columns. These polymer columns provide the required boundary condition and impose homeotropic alignment of LCs. The weak under developed micro-fibrils in the inter-column region further enhance the homeotropic director alignment.

The homeotropic alignment of LC induced by these polymer columns can be tentatively explained on the basis of anisotropic nematic elastic energy considerations proposed by Berreman.¹² Due to the narrow diameter of the polymer columns and planar anchoring on column walls, the LC director aligns parallel to the polymer walls (i.e., perpendicular to the substrate) to minimize the total elastic energy. Consequently, the cell becomes relatively free of director distortions. The effective LC anchoring energy of the resultant surface depends on the number density and size distribution of the polymer columns, which are determined by the physico-chemical nature and concentrations of the LC and pre-polymer, the cell thickness, and the dynamics of phase separation. In the devices, prepared by this method, the polymer columns may scatter light to an extent comparable to the spacers used in traditional LCD panels. The light scattering can be minimized by proper optimization of parameters, such as the exposure time, temperature, UV intensity, etc., to obtain well separated polymer columns spanning the cell thickness and bonding to both substrates without compromising the electro-optic performance of the device.

Figure 1(a) shows a scanning electron microscope (SEM) image (top view) of the polymer columns formed in a mixture of NOA65 and MJ, after the LC has been washed away with hexane. Figure 1(b) shows a tilted and magnified view of one of the columns. It is clear that the polymer columns extend vertically from one substrate to the other. In the region, indicated by A, an ultra-thin polymer layer covering the substrate, with micro-fibrils extending in the vertical direction, is formed.

Figure 2(a) shows microscopic LC textures of a 3 μm thick LC cell with glass substrates, using a mixture of

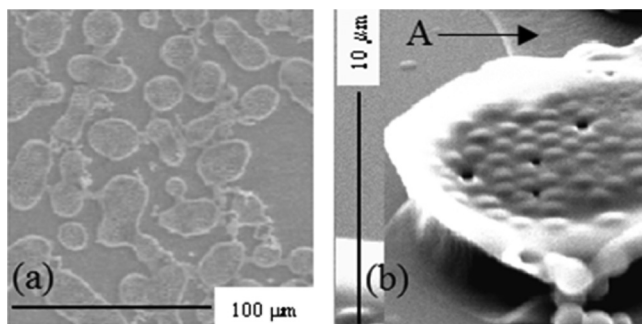


FIG. 1. The scanning electron microscope image of polymer columns (a) top wide view and (b) magnified oblique view. The region A has thin polymer layer with vertical micro-fibrils.

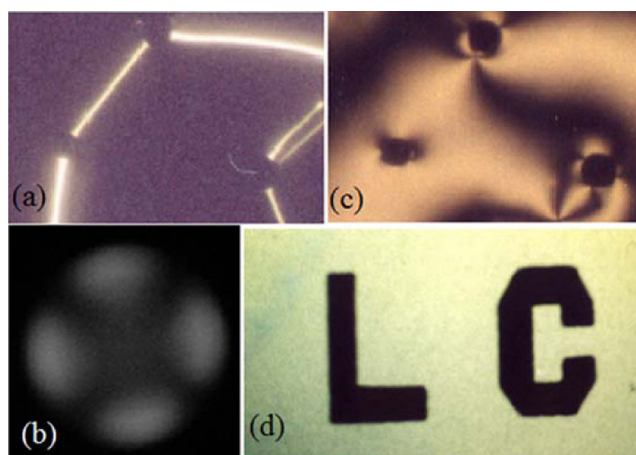


FIG. 2. (Color online) (a) Microscopic texture of a homeotropically aligned LC cell. (b) The conoscopic image confirming homeotropic alignment. (c) Microscopic texture after an external AC electric field is applied. (d) A prototype display on glass substrates. The difference in color in the background is the result of slight non uniformity in the cell thickness.

NOA68 and MJ (1:1 by weight), between crossed polarizers. The polymer columns appear as dark circles in Figures 2(a) and 2(c) between crossed polarizers with bright defect lines connecting them. Figure 2(b) shows a conoscopic image of the cell. The dark cross in the center of the field of view is invariant under the rotation of the cell about its surface normal, confirming homeotropic alignment of the LC director. Figure 2(c) shows microscopic texture of the cell after a square wave electric field of amplitude 1.33 V/ μm at 1 kHz is applied across the cell. Since the polymer columns are extended vertically with no tilt and the LC possesses negative dielectric anisotropy, the electric field reorients the LC director in the plane of the substrates with no preferential azimuthal orientation as indicated by the Schlieren texture. Figure 2(d) shows a prototype display (1 in. \times 2 in.) fabricated using 50 wt. % NOA68 in the mixture with glass substrates. The ITO electrodes are etched in the segmented

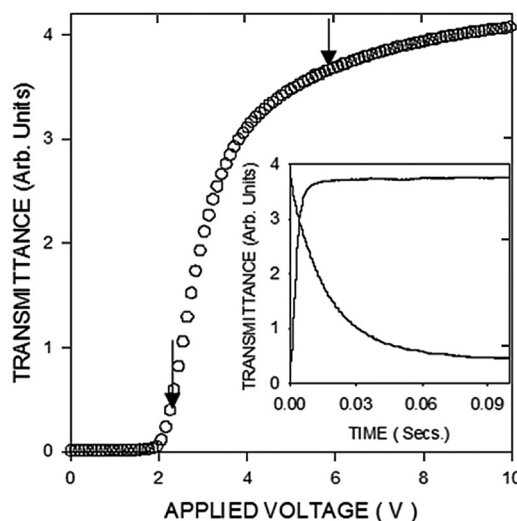


FIG. 3. Transmittance-voltage characteristics of a homeotropic cell filled with nematic LC possessing negative dielectric anisotropy. The vertical arrows indicate the voltages that correspond to 10% (OFF state) and 90% (ON state) of the maximum transmission. The inset shows the switching time characteristics of the same cell between the ON and OFF states.

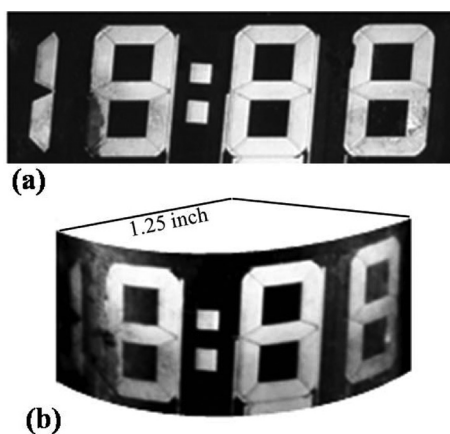


FIG. 4. A prototype alphanumeric 2 in. \times 3 in. flexible display made using polyethersulphone substrates (a) in flat geometry (no mechanical strain) (the lower left electrode segment of the first “8” is partially damaged) and (b) wrapped around a cylindrical object of 1.25 in. radius with its axis in the vertical direction.

(“LC”) area, so that the application of an electric field switches the remaining (the background) area of the cell.

Figure 3 shows the transmittance vs. voltage (T-V) plot for a $3\ \mu\text{m}$ thick LC cell fabricated with glass substrates using a 1:1 mixture of NOA68 and MJ. The gradual increase in the transmittance above $\sim 6\ \text{V}$ (marked with the vertical arrow) is due to anchoring of the LC on the wall of the polymer columns and a small amount of LC trapped inside the polymer columns. Inset in Fig. 3 shows the switching behavior of the same cell between ON and OFF states corresponding to 2.25 and 5.8 V, respectively. The response times of the cell measured between 10% and 90% of maximum transmittance (for both, turn-ON and turn-OFF states) are in the 10–50 ms range, which are typical values for nematic LC devices. These electro-optic behaviors are almost identical to those for conventional *vertical alignment* LC cells prepared using homeotropic alignment layers and LCs with negative dielectric anisotropy¹³ with the exception of the slow rise.

To demonstrate the applicability of the method for fabrication of flexible LC displays, a 2 in. \times 3 in. prototype display cell was fabricated using a mixture of NOA77 and MJ (in 1:1 weight ratio) and $100\ \mu\text{m}$ thick birefringence-free polyethersulphone substrates separated by $5\ \mu\text{m}$ spacers. Figure 4(a) shows the display in the ON state. Here, the active regions are the segments across which a square wave electric field of $1.2\ \text{V}/\mu\text{m}$ amplitude at 1 kHz is applied. Figure 4(b) shows the same display in the ON state but wrapped around a vertical cylinder of 1.25 in. radius. The cell appears grayish at the edges (where the viewing angle is approaching 90°)

due to non-zero optical birefringence offered to the transmitted light by the oblique orientation of the LC director. However, at and near normal view, the contrast ratio does not change due to flexing. Since the plastic substrates are bonded together at a fixed separation by polymer columns, the application of external pressure has minimal effect on the display, and it returns to its normal thickness/state almost immediately after the pressure is released.

In summary, we have demonstrated a simple one-step low-cost process to affect phase separation of LC-polymer mixture resulting in internal cell architecture consisting of polymer columns bonded to the two substrates. The anchoring conditions offered to LC by these columns and polymer micro-fibrils align the nematic director homeotropically. This mode is ideally suited for devices incorporating LC materials possessing negative dielectric anisotropy. This technology not only avoids the most undesirable process of rubbing alignment layers, but also eliminates several time consuming and costly steps currently employed by the LCD industry. The method presented here has the potential of being the basis of the next generation of flexible lightweight LCDs for the 21st century.

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