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Reduced operating voltage and grey-to-grey response time in a vertically aligned liquid crystal display using a mixture of two polyimide alignment materials

Ji-Hoon Lee¹, Young Eun Choi², Jun Hee Lee², Byeong Hoon Lee², Won Il Song³, Kwang-Un Jeong², Gi-Dong Lee^{3,4} and Seung Hee Lee^{2,4}

¹ Advanced Electronics and Information Research Center, Division of Electronic Engineering, Chonbuk National University, Jeonju, Jeonbuk 561-756, Korea

² Department of BIN Fusion Technology and Department of Polymer-Nano Science Technology,

Chonbuk National University, Jeonju, Jeonbuk 561-756, Korea

³ Department of Electronics Engineering, Dong-A University, Pusan 604-714, Korea

E-mail: gdlee@dau.ac.kr and lsh1@chonbuk.ac.kr

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Abstract

We proposed a method to reduce the operating voltage and the grey-to-grey switching time of a vertically aligned liquid crystal display using a mixture of planar and vertical polyimide alignment materials. The surface anchoring energy of the two-polyimide mixture was smaller than that of the pure vertical polyimide and consequently, liquid crystal molecules were easily switched to a planar state with an electric field, resulting in a greater maximum retardation than that of the pure polyimide at the same applied voltage. Rising time was also significantly reduced due to the suppressed optical bouncing effect in the mixed planar polyimide, and the decaying time showed negligible change. With the proposed approach, we can reduce the cell gap to obtain half-wave retardation allowing for faster response time while keeping a low operating voltage.

(Some figures may appear in colour only in the online journal)

1. Introduction

Various liquid crystal display (LCD) modes, such as vertical alignment (VA) [1–12], in-plane switching (IPS) [13], and fringe-field switching (FFS) [14–17], have been developed to improve the image quality of LCDs. The VA mode exhibits a high contrast ratio for normal viewing and can be fabricated simply without need for rubbing. Although the viewing angle properties of the VA mode are inferior to the other modes, this has been overcome using multi-domain technology with protrusion (MVA) [1–3], patterned electrodes (PVA) [4–6],

or fine electrodes with a polymer-stabilization (PS) technique (PS-VA) [7–12]. However, the large operating voltage and slow response time of the VA mode due to the small negative dielectric anisotropy and high rotational viscosity of the LC remain an open issue. In particular, the grey-to-grey response time of VA-LCDs is slow, typically above tens of milliseconds, and a special driving method, such as over-driving, is required. Generally, an increase in the negative dielectric anisotropy of LC accompanies an increase in rotational viscosity. Thus, varying LC parameters to decrease the operating voltage and to enhance the response time were not successful.

In this report, we suggest a method to reduce the operating voltage and the grey-to-grey response time of VA mode LCDs

⁴ Authors to whom any correspondence should be addressed.



Figure 1. Schematic illustration of the postulated LC orientation on the mixed alignment layer for (a) $p \gg v$, (b) $p \sim v$, (c) $p \ll v$.

using a mixture of planar and VA materials which have a smaller surface anchoring energy than the pure VA polyimide. Several methods to reduce the surface anchoring energy by stacking vertical and planar alignment layers [18-21] or cleaving off the side-chains of a VA polyimide [22] have been reported. However, these approaches require additional coatings or UV curing processes. To avoid additional processes, we used a method of mixing the vertical and planar alignment materials. Because the two polyimide materials have different chemical properties, they were phase separated to form vertical and planar domains with sizes smaller than a visible wavelength. Provided that the sizes of the planar domains are sufficiently small compared with the vertical domains, the overall vertical orientation of the LC molecules does not change, while the surface anchoring energy can be reduced. Under the weak surface anchoring condition, LC molecules were easily tilted by a smaller electric field and thus the maximum retardation at a given voltage was increased. In this situation, we could reduce the cell gap d to have a retardation of $\lambda/2$ compared with the strong anchoring cell. We measured the electro-optical response of the thinner cell with weak surface anchoring and found a significant reduction in the operating voltage and the grey-to-grey rising time. The change in the decay time was negligible. The suggested method has the benefit of requiring no additional fabrication process or special driving technique.

2. Experimental conditions

We prepared two kinds of cells; mono-domain cells with no electrode pattern and multi-domain cells with an electrode pattern. In the mono-domain cell, a mixture of the vertical and planar polyimide in weight ratios of 100:0, 70:30 and 55:45 was coated on two substrates and then baked at 230° C for 1 h. Then, both substrates were rubbed with a cotton cloth and assembled in an antiparallel configuration with a cell gap of $4.0 \,\mu$ m. The variance of the cell gap was less than $0.05 \,\mu$ m. Commercially available polyimide AL60702 (JSR) and SE6514 (Nissan) were used as the vertical and planar alignment materials. The two polyimides were mixed in the solvent, then phase separated after drying, forming domains smaller than a visible wavelength. The orientation of the LC

molecules was determined by the relative area ratio between the vertical and planar domains. An LC mixture with a negative dielectric anisotropy $\Delta \varepsilon = -4$ [Merck-Japan] was injected by capillary force at 80 °C. The birefringence Δn of the LC was 0.077 at 589.3 nm, the rotational viscosity was $\gamma_1 = 136$ mPa s, the splay and bend elastic constants were $K_{11} = 13.5$ pN and $K_{33} = 15.1$ pN.

For the multi-domain cell, the vertical polyimide was mixed with the planar polyimide in weight ratios of 100:0, 55:45 and 45:55, coated on the substrate with a patterned electrode, then baked at 230 °C for 1 h. The PS technique [7–11] was used instead of rubbing in the multi-domain cell to induce a uniform tilt of the LC molecules. The width and separation of the patterned electrode were 3 μ m and 4 μ m, respectively. The cell gap of the multi-domain cell was $3.2 \pm 0.05 \,\mu$ m. In the multi-domain cell, an LC mixture with $\Delta \varepsilon = -3.6$, n = 0.0886, $\gamma_1 = 110 \,\text{mPa}\,\text{s}$, $K_{11} = 11.8 \,\text{pN}$ and $K_{33} = 12.4 \,\text{pN}$ [MJ002430, Merck] was mixed with a 0.05 wt% reactive mesogen (RM257, Merck).

3. Results and discussion

Figure 1 shows a schematic illustration of the postulated LC orientation on the mixed alignment layer. Given the large diameter p of the planar polyimide coated zone, the LC molecules will be planar aligned forming a separate domain (figure 1(a)). If p is small but non-negligible compared with the diameter of the vertical domain v, the LC orientation is deformed and polar-horizontal or polar-vertical defects [23] will appear, depending on the pretilt angle of the neighbouring vertical domains (figure 1(b)). With very small p, comparable to the nematic correlation length, the LC molecules are vertically aligned with a reduced surface anchoring energy (figure 1(c)).

The weight fraction *R* of the vertical polyimide was defined as $R = [AL60702] / \{[AL60702] + [SE6514]\}$. Here, *R* represents the weight fraction of solid contents after solvent evaporation. We measured the transmittance of the cells with R = 1.0, 0.7 and 0.55 relative to the applied voltage across the cell (figure 2). To measure the voltage–transmittance response, collimated white light from a halogen lamp was passed through a polarizer, the cell, an analyser and a detector.



Figure 2. Voltage–transmittance curves of the mono-domain cells with R = 1.0, 0.7 and 0.55. A bipolar voltage with a frequency of 60 Hz was applied across the cell. Lines represent the simulated curve using the material parameters.



Figure 3. $(R/R_o - 1)(V - V')$ of the R = 1.0, 0.7 and 0.55 cells versus V - V'. The slope of the graph corresponds to $2K_{33}/Wd$.

The rubbing direction of the mono-domain cell was at 45° to the polarization direction of the incident light. The variance of the cell gap was less than $0.05 \,\mu$ m, and thus the variation of the retardation due to the cell gap was less than 4 nm at a wavelength of 550 nm. The R = 0.55 cell (green triangle) showed the maximum transmittance at \sim 4.7 V and the transmittance decreased at higher voltages. Thus, the retardation of the R = 0.55 cell was greater than $\lambda/2$. On the other hand, the transmittance of the R = 1.0 cell was not saturated until 10 V, indicating that the retardation of the cell was less than $\lambda/2$. We directly measured W of the fabricated cells (figure 3) by checking the change in retardation according to the method in the literature [24, 25]. Thicker cells with a cell gap of 10 μ m were used for the surface anchoring energy measurement under the same surface treatment conditions. As described in [24, 25], $(R/R_o - 1)(V - V')$ was plotted versus V - V', where R is the retardation under the given voltage, R_o is the maximum retardation under 50 V, and $V' = \sigma \beta V_{\text{th}}$ where $\sigma = (1/\pi) \int_{0}^{1} \sqrt{(1+\beta)(1+km)/(1+\beta m)m} \, dm$ with $\beta = (\varepsilon_{\perp}/\varepsilon_{ll}) - 1$ and $k = (K_{11}/K_{33}) - 1$. The fitted slope of the graph corresponds to $2K_{33}/Wd$. The cells with R = 1.0, 0.7, and 0.55 had values of W equal to 1.7×10^{-4} , 4.7×10^{-5} and 4.0×10^{-5} J m⁻². We also measured pretilt angle θ_p according to the method described in the literature [23, 26]. In cells with R = 1.0, 0.7 and 0.55, the values of θ_p were 87.5°, 87.3° and 87.3°. Using the material parameters including W and θ_p , we simulated the voltage–transmittance curve (lines in figure 2). Although there was a slight difference between the simulation and experiments, the simulated data were qualitatively approximated to the experimental data. Thus, the effect of the reduced surface anchoring on the voltage–transmittance data is confirmed.

The surface extrapolation length b = K/W converges to zero under an infinite surface anchoring, where K and Ware the elastic and surface anchoring coefficients, respectively. For cells with R = 1.0, 0.7 and 0.55, the values of b were 0.09, 0.32 and 0.37 μ m. These results indicate that we can reduce the cell gap by approximately $2b = 0.74 \,\mu\text{m}$ to obtain a $\lambda/2$ retardation with weak surface anchoring of $4.0 \times 10^{-5} \,\mathrm{J}\,\mathrm{m}^{-2}$. In addition, the Fréedericksz transition voltages of the cells with R = 1.0, 0.7 and 0.55 were 1.9, 1.8 and 1.7 V. The theoretical threshold voltage $(V_{\rm th})$ of an LC cell is given by $V_{\rm th} = E_{\rm th}d = \pi (d + 2b)^{-1} \{K/(\varepsilon_0 \varepsilon_a)\}^{1/2} d$ [22, 27]. The theoretically calculated values of V_{th} for cells with R = 1.0, 0.7and 0.55 cells were 1.94, 1.85 and 1.83 V, which qualitatively approximated the experimental values. It should also be noted that the transmittance was at 90% of the maximum at voltages of 5.0 V and 3.8 V in the cells with R = 1.0 and 0.55, respectively. Thus, the operating voltage was reduced using the mixture of planar and vertical polyimides.

To confirm the existence of domains with a different surface anchoring energy, we observed the polarizing optical microscopy (POM) image of the cells with R = 1.0 (figures 4(*a*) and (*b*)) and R = 0.55 (figures 4(*c*) and (*d*)) at 0 and 2.9 V applied. In the zero field state, both cells with R = 1.0 (figure 4(*a*)) and R = 0.55 (figure 4(*c*)) showed uniform dark states without domains. Thus, as in figure 1(*c*), the domain size of the planar alignment layer was small in the cell with R = 0.55, and the LC molecules were uniformly vertically oriented. Upon applying 2.9 V across the cell, the cell with R = 1.0 showed a uniform grey state (figure 4(*b*)), while the cell with R = 0.55 showed tiny domains with a brighter intensity (the circles in figure 4(*d*)). The bright domain in the mixed-polyimide cell indicates the existence of domains with smaller V_{th} due to the reduced surface anchoring energy.

A question could be raised about the absence of a planar domain in the zero field state in spite of the high concentration (45 wt%) of planar polyimide in the R = 0.55 cell. We think the more hydrophobic vertical polyimide segregated to the air interface and covered most of the surface, as reported in the literature [26, 28]. The surface free energy from the Landau model is given by $f(\theta) = \frac{1}{2}(A\sin^2\theta + B\cos^2\theta + C\sin^4\theta + D\sin^4\theta)$, where A and B are the quadratic anchoring coefficients, and C and D are quartic coefficients [26]. For small deviations from the planar ($\theta = 0^\circ$) and vertical orientations ($\theta = 90^\circ$), the Rapini–Papoular approximation considering only the quadratic terms is valid



Figure 4. POM image of the mono-domain cell: (a) R = 1.0 at 0 V, (b) R = 1.0 at 2.9 V, (c) R = 0.55 at 0 V and (d) R = 0.55 at 2.9 V. The white domains in (d) correspond to the planar domains with reduced V_{th} . Scale bars correspond to 50 μ m.



Figure 5. (*a*) Pixel electrode structure of the multi-domain cell. Width and separation of the electrodes were 3 μ m and 4 μ m, respectively. POM image of the polymer-stabilized multi-domain cells: (*b*) R = 1.0, (*c*) R = 0.55 and (*d*) R = 0.45. Scale bars correspond to 100 μ m.

and $f(\theta) = \frac{1}{2}B + \frac{1}{2}(A - B)\sin^2\theta$. Thus, a uniform vertical orientation can be stable given polyimide mixtures of A < B.

We fabricated multi-domain cells with a patterned electrode (figure 5(*a*)) and examined the optical response of the cells. To confirm the reduction in operating voltage and response time, we fabricated a multi-domain cell that was thinner than the mono-domain cell. The cell gap of the multi-domain cell was $3.2 \pm 0.05 \,\mu$ m. To optimize the retardation of the multi-domain cells to $\sim \lambda/2$, we used an LC mixture with n = 0.0886 (MJ002430 from Merck-Japan) which was slightly greater than that used in the mono-domain

cells. To induce pretilt of the LC molecules, we used the PS technique: a voltage much greater than V_{th} was applied across the cell and a UV light was used to polymerize the reactive mesogen molecules in the cell. The field-induced tilt of the LC molecules was fixed by the polymerized mesogen molecules.

Figure 5 shows the electrode structure and the POM images of the cells with R = 1.0 (figure 5(*b*)), R = 0.55 (figure 5(*c*)), and R = 0.45 (figure 5(*d*)) in the zero field state, 50% transmitting state, and 90% transmitting state. Similar to the mono-domain cell, the cell with a greater fraction of the planar polyimide had a more yellowish colour in the 90%



Figure 6. Transmittance versus applied voltage across the multi-domain cells with R = 1.0 (square), R = 0.55 (circle), and R = 0.45 (triangle) before (empty notation) and after UV exposure (solid notation). Lines are simulated data using the material parameters.

transmitting state. The yellowish colour indicates retardation over 275 nm. Thus, greater retardation of the cell with a higher fraction of planar polyimide was also confirmed in the multidomain cell. We should note that the brighter domains of the mono-domain cell in the grey state (figure 4(*b*)) were not observed in the multi-domain cell (figure 5). This is presumably due to the fine separation and width of the electrode pattern. The width and separation of the electrodes were 3 μ m and 4 μ m, respectively (figure 5(*a*)), which was comparable to the LC domain size, and thus the domain was not apparent.

We measured the voltage-transmittance response of the multi-domain cells with R = 1.0 (square), R = 0.55 (circle) and R = 0.45 (triangle) before and after UV exposure (figure 6). The electrode direction was at 45° to the polarization of the probe light. After UV exposure, the values of $V_{\rm th}$ for the cells decreased slightly. This was due to the pretilt angle effect of the polymerized cells. The values of θ_p for the multi-domain cells with R = 1.0, 0.55 and 0.45 were $88.2^{\circ}, 88.0^{\circ}$ and 87.9° . Similar to the mono-domain cells, the multi-domain cells also showed a greater retardation value with smaller R. The retardation of the R = 0.45 cell was $\lambda/2$ at 7.6 V, while the R = 1.0 cell showed a retardation of less than $\lambda/2$ until 8 V. Thus, the greater retardation and smaller operating voltage of the mixed-polyimide cell relative to the pure polyimide cell were also confirmed in the multi-domain cells. We also measured W for the multi-domain cells to be 2.0×10^{-4} , 5.8×10^{-5} and 4.7×10^{-5} J m⁻² for the cells with R = 1.0, 0.55 and 0.45. The W values increased slightly compared with the mono-domain cells, which was probably due to the different LCs used. The dashed line in figure 6 represents the simulated voltage-transmittance data using the experimental parameters. Commercial LCD simulator Techwiz 3.0 (Sanayi system) was used. The simulated data can be approximated to the experimental values, indicating the effect of the weak surface anchoring on the voltage-transmittance data.

To examine the response time of the cells, we observed the time evolution of the multi-domain cells with R = 1.0 (figure 7(*a*)), R = 0.55 (figure 7(*b*)) and R = 0.45 (figure 7(*c*)) before and after PS. The voltage which induces 90% transmittance of the cell was applied and the textures were captured in the course of LC switching. The elapsed time τ for texture stabilization with disappearance of disclination lines was very long at over a second before PS, and significantly decreased after PS in each cell. The value of τ for the cell with R = 1.0 was ~4000 ms before PS, and reduced to 82.1 ms after PS (figure 7(*a*)). On the other hand, τ after PS of the cells with R = 0.55 (figure 7(*b*)) and R = 0.45 (figure 7(*c*)) were 9.6 and 4.7 ms. Thus, the values of τ for the planar polyimide mixed cells were about an order of magnitude faster than the cell with R = 1.0.

Figure 8 shows the rising and decaying time of the multi-domain cells versus greyscale. The greyscale value was defined as the normalized transmittance divided by the maximum transmittance showing a $\lambda/2$ retardation. The rising time τ_r and decaying time τ_d were defined as the time elapsed between a 10% to a 90% transmitting state and vice versa. The time τ_r of the PS cells (solid notation) was much faster than that of the un-polymerized cells (empty notation), and was further reduced for smaller values of R (figure 8(a)). In other words, $\tau_{\rm r}$ decreased with a greater fraction of the planar polyimide. We note that τ_r of the PS cells with R = 0.55 and R = 0.45was smaller than half of τ_r for the R = 1.0 cell. The value of τ_d increased with greater fractions of the planar polyimide (figure 8(*b*)). However, the difference in τ_d between the cells was less than 1 ms. Note the difference in the vertical scales of figures 8(a) and (b). Therefore, the fraction of the planar polyimide had little influence on τ_d .

Using the effective cell gap method [27], τ_r and τ_d of the VA LC cells are given by, $\tau_{\rm r} \approx \gamma / \{\varepsilon_0 \varepsilon_{\rm a} E^2 - \pi^2 K / (d^2 +$ 4dK/W, $\tau_{\rm d} \approx (\gamma/K\pi^2)(d^2 + 4dK/W)$. Using the material parameters of MJ002430 LC, the calculated τ_r of the R = 1.0cell under a 1 V μ m⁻¹ electric field was 5.2 ms, while τ_r of the R = 0.45 cell was 4.8 ms, where $d = 3.2 \,\mu$ m. The values of W for the cells with R = 1.0 and R = 4.5 were 2.0×10^{-4} J m⁻² and 4.7×10^{-5} J m⁻², respectively. Thus, the predicted value of τ_r for the weak anchoring cell was slightly faster than for the strong anchoring cell. However, the measured value of τ_r was significantly reduced for greater values of R (figure 8(a)). To understand this abnormal behaviour, we examined the time evolution of the transmittance in the presence of the electric field. Figures 8(c) and (d) are oscilloscope images of transmittance versus time upon application of 6.8 V across the cells with R = 1.0 and R = 0.45, respectively. An optical bouncing phenomenon was observed around $t = 12.5 \,\mathrm{ms}$ (figure 8(c)) and was due to the LC molecules switching to opposite directions, $+\theta$ or $-\theta$. This explains the long τ_r (figure 8(a)) compared with τ_d (figure 8(b)). Meanwhile, the optical bouncing effect was significantly removed in of the cell with R = 0.45 (figure 8(*d*)). We should note that the same LC and cell gap were used in the R = 1.0 and 0.45 multi-domain cells. Thus, the reduced optical bouncing effect is not related to the bulk effect. Presumably, the mixed planar alignment material promoted the uniform pretilt of the LC molecules, and this is thought to be related to the reduction in bouncing effect. On the other hand, the calculated values of τ_d for the strong



Figure 7. Time evolution of the POM textures of the multi-domain cells: (a) R = 1.0, (b) R = 0.55 and (c) R = 0.45 under the applied voltage across the cell. The voltage inducing a 90% transmittance was applied and the textures were captured.

and weak surface anchoring cells were 12.0 ms and 15.6 ms, respectively. However, the difference between the measured values of τ_d for the strong and weak surface anchoring cells was less than 1 ms, as mentioned above. We think this may be related to the uniform tilt of the LC molecules caused by the planar polyimide, reducing a nonlinear internal flow effect.

4. Conclusions

We proposed a method to reduce the operating voltage and grey-to-grey switching time of a vertically aligned liquid crystal display using a vertical and planar polyimide mixture. The planar polyimide reduced the vertical surface anchoring energy, and greater retardation was obtained with a smaller voltage. Then, we could reduce the cell gap by $\sim 2b$ to

achieve a $\lambda/2$ retardation. The switching voltage of the planar polyimide mixed cell was smaller than that of the pure vertical polyimide cell. The rising time was significantly reduced by the diminished optical bouncing effect, while the decaying time was nearly unchanged. The method suggested in this paper can reduce the operating voltage and grey-to-grey response time of VA-LCDs without requiring an additional fabrication process or special driving technique.

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Figure 8. (*a*) Rising time and (*b*) decaying time of the multi-domain cells versus greyscale. Captured oscilloscope image of the transmittance versus time upon applying 5.7 V across the cell: (*c*) R = 1.0 and (*d*) R = 0.45.

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