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Flexible Liquid Crystal Displays Using Liquid Crystal-polymer Composite Film and Colorless Polyimide Substrate

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Application of liquid crystal (LC) materials to a flexible device is challenging because the bending of LC displays easily causes change in thickness of the LC layer and orientation of LCs, resulting in deterioration in a displayed image quality. In this work, we demonstrate a prototype device combining a flexible polymer substrate and an optically isotropic LC-polymer composite in which the device consists of interdigitated in-plane switching electrodes deposited on a flexible colorless polyimide substrate and the composite consisting of nano-sized LC droplets in a polymer matrix. The device can keep good electro-optic characteristics even when it is in a bending state because the LC orientation is not disturbed in both voltage-off and -on states. The proposed device shows a high potential to be applicable for future flexible LC devices.

Keywords : Flexibility, Optically isotropic liquid crystal (OILC), Colorless polyimide (CPI), In-plane switching (IPS)
OCIS codes : (230.2090) Electro-optical devices; (230.3720) Liquid-crystal devices

I. INTRODUCTION

Flexible displays receive great attention due to their advantages such as their light weight, thin packaging, impact resistance, and lack of a spatial limit of use compared with conventional glass-based displays [1, 2]. Many different types of flexible displays utilizing liquid crystal displays (LCDs) [3], organic light emitting diodes (OLEDs) [4, 5], light emitting diodes (LED) [6] and particle-used electrophoretic displays [7] have already been demonstrated for wearable displays, smart cards and electronic papers. At present, LCDs with various driving modes, such as twisted nematic (TN), vertical alignment (VA) [8, 9] in-plane switching (IPS) [10], and fringe-field switching (FFS) [11-13], have been commercialized, dominating most of the display markets with high image quality. The present LCDs utilize glass substrates because they can preserve orientation and thickness of LCs and it is advantageous to perform the manufacturing process in a large-size area. On the other hand, plastic-based OLED is commercialized although the manufacturing process is performed with a glass substrate so that the display can be made with curved form, lightweight and thinness. To overcome intrinsic demerits of the glass-based LCDs, the

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plastic substrate-based FFS-LCDs were recently developed by Japan Display Inc. [11-13], whose LCDs are lighter, thinner, and have smaller curvature. However, the LCD has some limitations in bending and mechanical stability because the bending may cause perturbation of LC orientation and cell gap change, and external mechanical shocks may easily break LC orientation, resulting in deterioration of the image quality of LCDs and external mechanical shocks may easily break LC orientation. To address the above concerns plausible approaches were proposed, which build polymer walls between pixels by polymerization induced phase separation. Such relatively rigid parts of polymer walls can help to maintain the cell gap, they are expected to prevent flow of LCs under external pressure [1, 2, 14-17]. However, the issue is not fully solved by using such walls because the deformation of substrates can also induce the distortion against the uniform alignment of LC directors, which causes an optical leakage that gives rise to a bad effect for a dark state of LCDs. Further, it is not a fundamental solution to avoid the flow of LCs in the device. To solve these issues, several reports suggested LC-polymer composites in which nano-size LC droplets are embedded in the polymer matrix so that the composite becomes polarization independent optically isotropic liquid crystal (OILC) [16-20]. In this way, the flowing property of the LC layer disappears and the OILC film can bend more easily without changing its optical properties.

In this paper, we demonstrate a real flexible LCD in which a single substrate utilizing colorless polyimide (CPI) with interdigitated indium-tin-oxide (ITO) electrodes for in-plane switching (IPS) is used and LC-polymer composite is coated above the substrate. Since the composite is optically isotropic and CPI has no in-plane retardation, the cell shows a complete dark state under crossed polarizers. The electro-optic characteristics of the proposed flexible LCD in bending state were studied in detail.

II. EXPERIMENTAL

2.1. Fabrication of Flexible IPS-CPI Electrode Film

The flexible CPI film with patterned IPS electrode was fabricated in 6 steps. i) The CPI varnish was coated on the cleaned Si substrate by spin-coating under 1,000 rpm for 60 s, and it was cured at 60°C, 80°C, 150°C, 230°C, and 300°C for 30 min at each temperature in N_2 environment, ii) Deposition of an ITO layer on the CPI film was carried out by the RF-magnetron sputtering system (SDP-670VT, ULVAC), iii) Photoresist (PR) was coated on the ITO, iv) The photoresist development and bake proceeded to make a patterned ITO electrode, v) The exposed ITO was etched away by a prepared etching solution and residual PR was removed. vi) Finally, the fabricated IPS-CPI electrode film was peeled off from the Si substrate. The detailed process of preparing IPS-CPI with this photo-lithography technique is schematically shown in Fig. 1. To measure the thickness of the CPI film and ITO electrode, we used surface profiler (P-10, KLA-Tencor Corporation).

2.2. Fabrication and Characterization of Flexible OILC Cell

To prepare the OILC mixture, we used high dielectric anisotropic nematic LC mixture, MLC-2053 ($\Delta\varepsilon = 46.2$, $n_e = 1.7472$, $n_o = 1.5122$, $\Delta n = 0.235$ at 589.3 nm, from Merck Advanced Technology), UV-curable monomer, Norland Optical Adhesive 65 (NOA65, $n_p = 1.524$, from Norland Products Inc., USA), and a photo-initiator (Ciba, Irgacure651). The OILC mixture consists of 39.76 wt% of LC, 59.64 wt% of NOA65, and 0.6 wt% of photo-initiator. The prepared OILC mixture was coated on the CPI-IPS films. After coating, the mixture was cured by UV exposure under 150 mW/cm^2 for 5 s at room temperature, resulting in thickness of about 10 μm. The polarizing optical microscope (Nikon, ECLIPSE E600) was used to

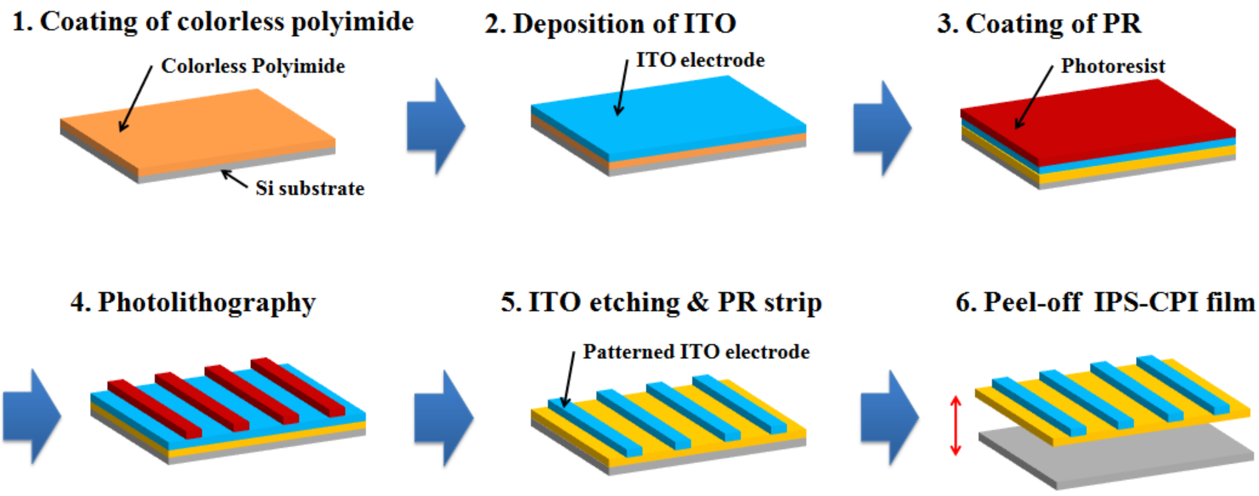

FIG. 1. Procedure for fabrication of the flexible in-plane electrode on the colorless polyimide (CPI) film.

observe the electro-optic properties of OILC in voltage on and off states. The length and width of patterned ITO electrode were measured by using scanning electron microscope (SEM, JSM-5900, JEOL). The voltage-dependent transmittance was measured by lab made set up with crossed polarizers, laser source (He-Ne laser, λ = 632.8 nm), a photo-detector, an oscilloscope (Tektronix, DPO2024B), an amplifier (FLC A400), and a function generator (Agilent, 33521A).

III. RESULTS

Generally, ITO is a very weak material against external stresses such as bending, stretching, and vibration. Nevertheless, the thinner substrates with deposited ITO can give better stability, flexibility, and longer lifetime under such stresses [21, 22]. To improve the stability and flexibility of patterned ITO electrode on CPI film, we fabricated a very thin CPI film with thickness of 13.5 μm and patterned ITO electrode with thickness of 108 nm in which the interdigitated electrode was patterned with electrode width of 4.24 μm and spacing of 5.60 μm between them to generate in-plane electric field in voltage-on state, as shown in Fig. 2.

The schematic of switching principle of the in-plane field driven OILC device using the CPI film under crossed polarizers is shown in Fig. 3. When the LC droplet size in a polymer matrix is roughly similar to the wavelength of incident light, it looks quite opaque due to high scattering. However, when its size is much smaller than the wavelength of the incident light, less than 250 nm, the scattering is minimized, resulting in a transparent optically isotropic phase [23, 24]. In the proposed OILC film, the LC droplet size is in a range of 100 to 200 nm so that the LC/polymer composite film shows high transparency. Consequently, it appears dark under the crossed polarizers, as shown in Fig. 3(a) and when a voltage is applied, the in-plane electric fields are formed so that the LCs inside droplets orient along the field direction, giving rise to field-induced birefringence known as the Kerr effect [25]. Therefore, the device gives rise to a bright state, when the optic axis of the induced birefringence makes an angle of 45° with respect to the crossed polarizer axes, as shown in Fig. 3(b). To measure the electro-optic properties, the OILC film is fixed between the crossed polarizers in such

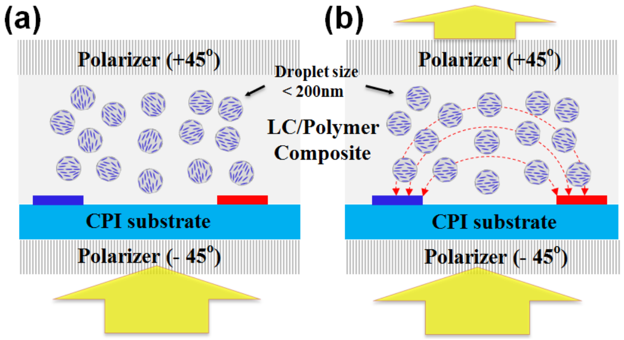

FIG. 3. Schematic of the flexible optically isotropic liquid crystals (a) at voltage-off and (b) voltage-on states.

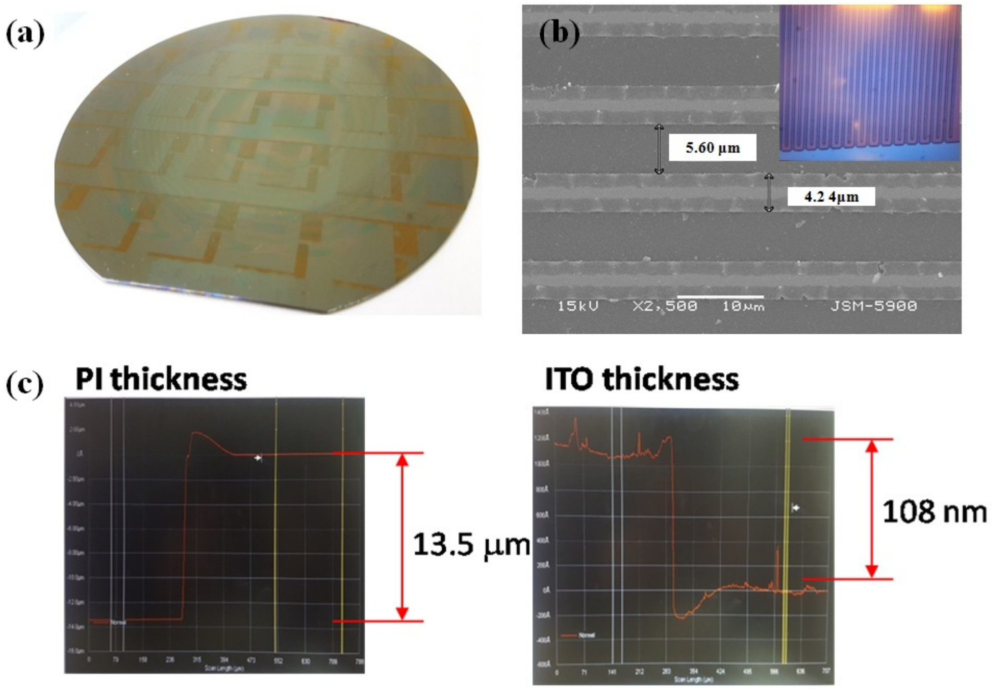

FIG. 2. (a) Thickness of CPI film and patterned ITO electrode layer, (b) SEM and OM image (inset) of patterned ITO electrode, and (c) photograph of IPS-CPI electrode film.

a way that the long-side electrode direction is 45° to the polarizers. The He-Ne laser light is made incident on the cell and the transmitted intensity detected and measured by using the photo-diode and the digital oscilloscope while applying 1 kHz frequency square wave voltage to the cell.

The Fig. 4 shows voltage-dependent transmittance of prepared OILC with flexible IPS substrate. The two inset images show the macroscopic images of black and bright states in bending state, respectively. No transmitted intensity was observed at 0 V, indicating that there is no light leakage from the film. When applying voltage, the transmittance was increased with the increase of voltage, finally reached to a maximum transmittance at 3.67 V/μm, indicating the induced birefringence is saturated at high enough electric field. The threshold electric field (E_{th}) and operating electric field (E_{op}) are defined as the voltage required achieving 10% and 90% transmittance relative to maximum transmittance, respectively. The measured E_{op} and E_{th} are 2.37 V/μm and 0.78 V/μm, respectively.

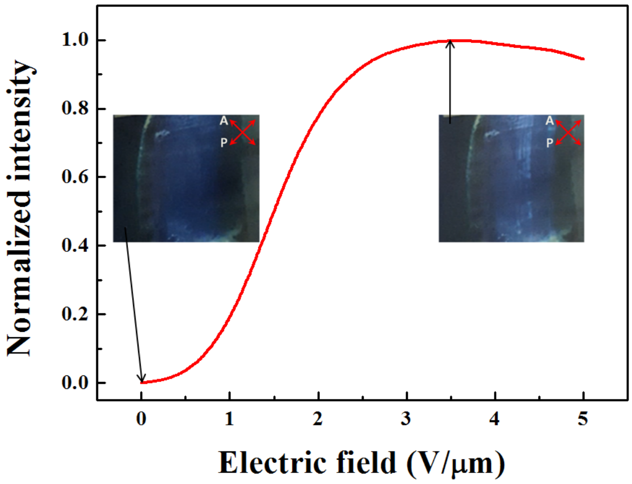

FIG. 4. The dependence of the normalized intensity of the flexible OILC cell with applied E-field. Two insets are photographs of OILC cell in the flat state taken at voltage-off (0 V/μm) and voltage-on (3.67 V/μm) states.

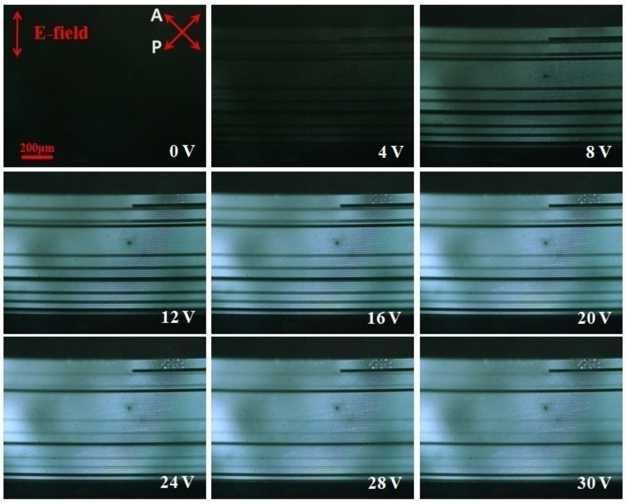

FIG. 5. The POM images of the flexible OILC cell taken at different bias voltages under crossed polarizers.

We also observed switching behavior of prepared cells through polarizing optical microscopy, as shown in Fig. 5. The aforementioned square wave field is applied to the cell to observe the switching behavior. As expected, the obtained film shows a dark state at the field off state. In addition, there is no indication of change in color on rotation of the cell under crossed polarizers, suggesting the obtained film is optically isotropic. Upon applying the voltage, the dark state start to give rise to brightness, which is indication of induced birefringence in the OILC film. The highest brightness was noticed at 3.67 V/μm. A long bright strip indicates the gap between the electrodes while the dark area indicates the area occupied by the electrodes. One could easily notice few either continuous or discontinues dark lines, in which the switching does not occur due to a few broken IPS electrodes.

Figure 6 shows set up of bending the film and the flexed OILC film with 9 mm of curvature [see Fig. 6(a)] and observed a distinct switching of nano-sized LC droplets from dark to bright state in top view under the cross polarizers. Under mechanical deformation, the LC orientation as well as the dark [see Fig. 6(b)] and white [see Fig. 6(c)] images are not disturbed while keeping an excellent uniformity, confirming an excellent cell gap is kept in the flexible OILC film using CPI film. However, the birefringence in the bright state was not shown over a whole area not because of bending state but due to broken IPS electrodes because of low quality photolithography process on a lab scale.

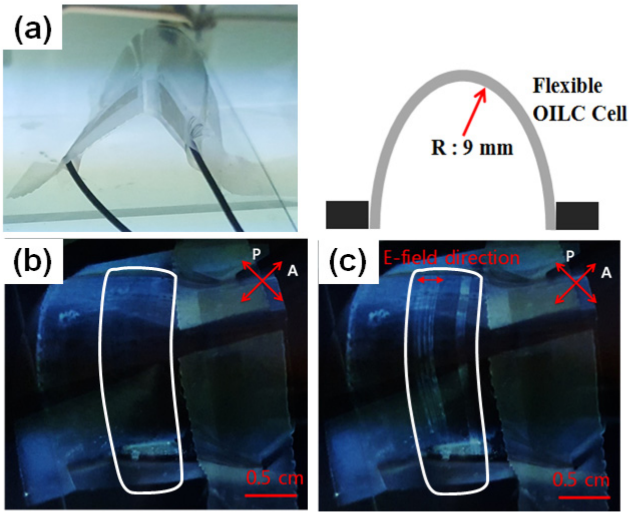

FIG. 6. (a) Photograph and schematic illustration of the flexible OILC cell in the bent state. (b) and (c) top view of OILC cell in OFF and ON state under cross polarizers, respectively.

IV. CONCLUSION

In this work, we demonstrate a fully flexible liquid crystal device with thickness of less than 30 μm in which a colorless polyimide is used as thin substrate and optically isotropic liquid crystal/polymer composite film is formed via photo-induced polymerization induced phase separation. The proposed device is fully flexible because its dark and white states are not disturbed by bending owing to captured LC in a polymer matrix and very light weight because the device is just polymer film. Although polarizers still remain as a quite big obstacle for the application to a flexible display, there are also good approaches for pursuing the flexibility of polarizers as well, such as coatable polarizers [26, 27]. It is believed that our approach can greatly contribute toward the application of LCs to flexible displays.

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REFERENCES

1. H. Sato, H. Fujikake, Y. Iino, M. Kawakita, and H. Kikuchi, "Flexible grayscale ferroelectric liquid crystal device containing polymer walls and networks," Jpn. J. Appl. Phys. **41**, 5302-5306 (2002).
2. H. Fujikake, H. Sato, and T. Murashige, "Polymer-stabilized ferroelectric liquid crystal for flexible displays," Displays **25**, 3-8 (2004).
3. D.-W. Kim, C.-J. Yu, Y.-W. Lim, J.-H. Na, and S.-D. Lee, "Mechanical stability of a flexible ferroelectric liquid crystal display with a periodic array of columnar spacers," Appl. Phys. Lett. **87**, 051017 (2005).
4. Y. Li, L.-W. Tan, X.-T. Hao, K. S. Ong, F. R. Zhu, and L.-S. Hung, "Flexible top-emitting electroluminescent devices on polyethylene terephthalate substrates," Appl. Phys. Lett. **86**, 153508 (2005).
5. A. N. Krasnov, "High-contrast organic light-emitting diodes on flexible substrates," Appl. Phys. Lett. **80**, 3853-3855 (2002).
6. M. Choi, B. Jang, W. Lee, S. Lee, T. W. Kim, H. J. Lee, J. H. Kim, and J. H. Ahn, "Stretchable active matrix inorganic light-emitting diode display enabled by overlay-aligned roll-transfer printing," Adv. Funct. Mater. **27**, 1606005 (2017).
7. A. K. Srivastava, M. Kim, S. M. Kim, M.-K. Kim, K. Lee, Y. H. Lee, M.-H. Lee, and S. H. Lee, "Dielectrophoretic and electrophoretic force analysis of colloidal fullerenes in a nematic liquid-crystal medium," Phys. Rev. E **80**, 051702 (2009).
8. S. G. Kim, S. M. Kim, Y. S. Kim, H. K. Lee, S. H. Lee, G. D. Lee, J. J. Lyu, and K. H. Kim, "Stabilization of the liquid crystal director in the patterned vertical alignment mode through formation of pretilt angle by reactive mesogen," Appl. Phys. Lett. **90**, 261910 (2007).
9. Y. J. Lim, H. J. Kim, Y. C. Chae, G. Murali, J. H. Lee, B. Mun, D. Y. Gwon, G. Lee, and S. H. Lee, "Fast switching and low operating vertical alignment liquid crystal display with 3-D polymer network for flexible display," IEEE Trans. Electron Devices **64**, 1083-1087 (2017).
10. M. Ohe and K. Kondo, "Electro-optical characteristics and switching behavior of the in-plane switching mode," Appl. Phys. Lett. **67**, 3895-3897 (1995).
11. S. H. Lee, S. L. Lee, and H. Y. Kim, "Electro-optic characteristics and switching principle of a nematic liquid crystal cell controlled by fringe-field switching," Appl. Phys. Lett. **73**, 2881-2883 (1998).
12. O. Shinichiro, H. Yosuke, J. Lu, Y. Yohei, A. Genki, H. Emi, H. Toshiyuki, and K. Shinichi, "58-1: Invited Paper: High resolution IPS-LCDs fabricated with transparent polyimide substrates," SID Symposium Digest of Technical Papers **49**, 764-767 (2018).
13. M. Kim, H. S. Jin, S. J. Lee, Y.-H. Shin, H. G. Ham, D.-K. Yang, P. J. Bos, J. H. Lee, and S. H. Lee, "Liquid crystals for superior electro-optic performance display device with power-saving mode," Adv. Opt. Mater. **6**, 1800022 (2018).
14. T. Murashige, H. Fujikake, H. Sato, H. Kikuchi, T. Kurita, and F. Sato, "Polymer wall formation using liquid-crystal/polymer phase separation induced on patterned polyimide films," Jpn. J. Appl. Phys. **43**, L1578-L1580 (2004).
15. P. Mach, S. J. Rodriguez, R. Nortrup, P. Wiltzius, and J. A. Rogers, "Monolithically integrated, flexible display of polymer-dispersed liquid crystal driven by rubber-stamped organic thin-film transistors," Appl. Phys. Lett. **78**, 3592-3594 (2001).
16. S. Matsumoto, M. Houlbert, T. Hayashi, and K. Kubodera, "Fine droplets of liquid crystals in a transparent polymer and their response to an electric field," Appl. Phys. Lett. **69**, 1044-1046 (1996).
17. Y. Tanabe, H. Furue, and J. Hatano, "Optically isotropic liquid crystals with microsized domains," Mater. Sci. Eng. B-Solid. **120**, 41-44 (2005).
18. S. W. Choi, S. I. Yamamoto, T. Iwata, and H. Kikuchi, "Optically isotropic liquid crystal composite incorporating in-plane electric field geometry," J. Phys. D: Appl. Phys. **42**, 112002 (2009).
19. M. Kim, B. G. Kang, M. S. Kim, M. K. Kim, P. Kuar, M. H. Lee, S. W. Kang, and S. H. Lee, "Measurement of local retardation in optically isotropic liquid crystal devices driven by in-plane electric field," Curr. Appl. Phys. **10**, E118-E121 (2010).
20. N. H. Park, S. C. Noh, P. Nayek, M.-H. Lee, M. S. Kim, L.-C. Chien, J. H. Lee, B. K. Kim, and S. H. Lee, "Optically isotropic liquid crystal mixtures and their application to high-performance liquid crystal devices," Liq. Cryst. **42**, 530-536 (2015).
21. K. D. Harris, A. L. Elias, and H. J. Chung, "Flexible electronics under strain: a review of mechanical characterization and durability enhancement strategies," J. Mater. Sci. **51**, 2772-2805 (2016).

22. C.-C. Lee, P.-C. Huang, and K.-S. Wang, "Flexural capability of patterned transparent conductive substrate by performing electrical measurements and stress simulations," Materials **9**, 850 (2016).
23. Y. J. Lim, J. H. Yoon, H. S. Yoo, S. M. Song, R. Manda, S. Pagidi, M.-H. Lee, J.-M. Myong, and S. H. Lee, "Fast switchable field-induced optical birefringence in highly transparent polymer-liquid crystal composite," Opt. Mater. Express **8**, 3698-3707 (2018).
24. S. Pagidi, R. Manda, Y. J. Lim, S. M. Song, H. S. Yoo, J. H. Woo, Y.-H. Lin, and S. H. Lee, "Hellical pitch-dependent electro-optics of optically high transparent nano-phase separated liquid crystals," Opt. Express **26**, 27368-27380 (2018)
25. C. M. Chang, Y. H. Lin, V. Reshetnyak, C. H. Park, R. Manda, and S. H. Lee, "Origins of Kerr phase and orientational phase in polymer-dispersed liquid crystals," Opt. Express **25**, 19807-19821 (2017).
26. P. Su, G. Vladmir, and S. K. Hoi, "P-155: High-performance coatable polarizer by photoalignment," SID Symposium Digest of Technical Papers **48**, 1866-1868 (2017).
27. S. K. Park, S. E. Kim, D. Y. Kim, S. W. Kang, S. Shin, S. W. Kuo, S. H. Hwang, S. H Lee, M. H. Lee, and K. U. Jeong, "Polymer-stabilized chromonic liquid-crystalline polarizer," Adv. Funct. Mater. **21**, 2129-2139 (2011).