

Optical Property Enhancement of Dye-PDLC Using Active Reflector Structure

Jae Eun Jung,¹ Gae Hwang Lee,¹ Jae Eun Jang,² Kyu Young Hwang,¹ Farzana Ahmad,³ Muhammad Jamil,⁴ Lee Jin Woo,³ Young Jae Jeon³

¹Display Lab: Materials & Device Laboratory, Samsung Advanced Institute of Technology, San 14, Nongseo-dong, Giheung-gu, Yongin-si, Gyeonggi-do 446-712, Korea

²Department of Information and Communication Engineering, Daegu Gyeongbuk Institute of Science and Technology (DGIST), Daegu 711-873, Korea

³Department of Chemistry, Liquid Crystal Research Center, Konkuk University, Seoul 143-701, Korea

⁴Division of International Studies, University College, Konkuk University, Seoul 143-701, Korea

Received 27 December 2010; accepted 30 May 2011

DOI 10.1002/app.35011

Published online 11 October 2011 in Wiley Online Library (wileyonlinelibrary.com).

ABSTRACT: The enhancement of electro-optical properties in reflective-type dye polymer dispersed liquid crystal (PDLC) has been achieved by applying an additional white PDLC layer along with dye PDLC layer. This newly modified structure that consists of white PDLC layer and scattering reflector acts as an active reflector. In this practice, an additional arrangement of a polymer barrier layer is made-up over white PDLC layer, to block the absorption of any solution from dye PDLC. The contrast ratio of this new configuration is almost

doubled with low driving voltage and high ON reflectance. Simultaneously, the "off" state has been observed darker than single layer dye PDLC structure. This new configuration can be potentially significant for various display applications such as E-paper, outdoor billboard, and flexible display. © 2011 Wiley Periodicals, Inc. *J Appl Polym Sci* 124: 873–877, 2012

Key words: polymer dispersed liquid crystal; optical properties; liquid crystal and polymer; dye-PDLC

INTRODUCTION

Polymer dispersed liquid crystals (PDLCs), representing an important new class of materials, which do not require polarizer, have potential applications as optical display devices.^{1–6} PDLCs are composite materials that consist of micron-sized liquid crystal droplets randomly distributed in the polymer matrix, formed by phase separation of the initially homogeneous liquid crystal-polymer mixture.⁷ Additional advantages of PDLC devices are; they show quick electro-optical response, no leakage of materials, low cost production, and simple fabrication process compared to other technologies like LCDs.^{8–10} As a result, PDLCs films have promising applications for the large-area display, flexible displays, the fabrication of reflective and projection displays, windows shutters, and holographic recording medias.^{7,10–15}

Generally, the inhomogeneous polymer/LC composites in PDLCs have a spatially varying refractive index and efficiently scatter light, which makes them turbid appearance. This turbid appearance can be darkened by doping the PDLC with a dichroic dye (guest-host configuration; referred as GH). Light

scattering state is changed simply by applying an electric field across the film to orientate the LCs droplet (and dye molecules) directors with field direction. In such state, at certain electric field amplitude, the film becomes transparent if the ordinary refractive index of the droplets matches the refractive index of the polymer.

In display and light switch applications, the relevant properties of PDLC films are the ON- (and threshold) voltage and the contrast ratio (CR). For practical reasons, the ON-state has to be achieved at low voltage, and the CR has to be as large as possible. Earlier, many groups have reported several studies for the enhancement of CR (transmission or reflective mode) and electro-optical properties of the liquid crystal polymer-dispersed films.^{7,16–20} However, this area still needs to be explored further.

One suitable solution can be the mixing of dichroic dye to PDLC. Because the dichroic dye has an optical anisotropy, the configuration of dichroic dye in PDLC affects positively to improve the optical property in dye-doped PDLC.²¹ When applied, field aligns liquid crystals and dichroic dyes with the direction of electrical field, and the refractive index mismatching and the light absorption by dyes are minimized, so that the light mostly transmits through the material. However, there are some problems involved with dye-doped PDLC as well. Because the CR evaluated with such films is not

Correspondence to: Y. Jae Jeon (yjjeon@konkuk.ac.kr).

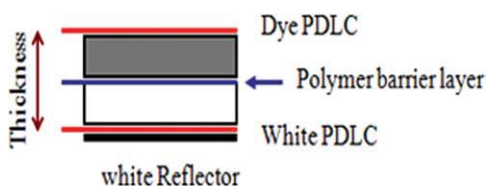


Figure 1 A schematic view of the double-layered PDLC with additional barrier layer. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

good enough. There are some reasons that cause such behavior. First, is the solubility problem which means that some dyes are dissolved not only in liquid crystal host but also in the polymer matrix. The dyes dissolved in the polymer matrix affect the light scattering and also change the absorbance. Second, the order parameter of dye is not as good as liquid crystal molecules, and the third is the low dye concentration.

The current work is an attempt to improve the electro-optical properties of dye-doped PDLC display. We proposed a new technique by inserting additional white PDLC layer along with the dye PDLC film. A UV-curable polymer layer was formed between these two PDLC films, which acted as one central wall between two PDLC layers (optically in series); all these films were connected electrically parallel; which greatly enhanced the CR of the device. The double-layered cells that consist of two PDLC films with a common polymer-based central wall have a function such as active reflector layer. We demonstrate that the total CR of double-layered cells working in reflective mode has been almost doubled of the individual dye PDLC cell CR with similar white reflectance value.

The device configuration and principle of our proposed PDLC device are shown in Figures 1 and 2. Our obtained results are discussed in Results and Discussion section. In the last section, conclusions of present studies have been described.

EXPERIMENTAL

Materials and methods

The assembly of our constructed reflective mode dye-doped PDLC is shown in Figure 1. To construct the reflective type white and dye-doped PDLCs, the following method has been used.

The dye-doped PDLC film was prepared by using an acrylate-based monomer (PN393, Merck), a liquid crystal (TL203, Merck), and a black azo type dye from G-series (Hayasibara Co., Japan). The prepolymer PN393 is commercially available; consists of a monomer, a crosslinker, a photoinitiator, and resin.

2-Ethylhexyl acrylate was used as the monomer, and Darocur4265 (Ciba) was used as a photoinitiator and trimethylolpropane triacrylate was used as the crosslinker in PN393,²² whereas the LC commercially available as TL203 is a mixture of pentyl cyano biphenyl blend with fluoro and chloro-substituted mesogens.¹⁰ First, a mixture composed of 20 wt % of the PN393 prepolymer and 80 wt % of the TL203 liquid crystal was prepared. Afterward, dye was added to this mixture with different weight ratios for the preparation of the sample dye-doped PDLC solution. The mixture was mechanically stirred well until it became homogenous. To define the gap of the future PDLC cell, mylar spacers of varying diameters size were used to maintain the cell gap.

The white PDLC film was prepared by using the same composition and materials as described earlier except with dye. To define the gap of the future PDLC cell, mylar spacers 10 and 20 μm in diameter were used to maintain the cell gap. For the construction of this double-layered PDLC, cell lift-off technique was used, the details of which are described elsewhere.²³

The polymer barrier (abbreviated as PB) layer between the white and dye PDLC films was prepared by a mixture of 1 : 1 ratio of highly viscous UV-curable polymer (provided by BASF) and isopropyl alcohol (IPA). For the purpose, spin-coating technique was used. To prepare the film, the spin coater with 500 rpm speed for 30 min was used and left this film at room temperature condition for complete evaporation of solvent.

The purpose to use this curable polymer coating as PB layer was to make a barrier layer between the

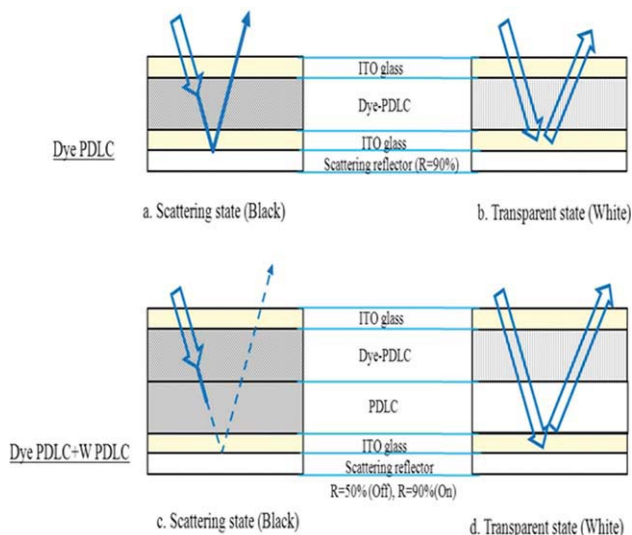


Figure 2 Principle and device configuration of single-layer dye-PDLC and current white-dye PDLC (W-DPDL). [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

dye and white PDLC to avoid the diffusion of dye molecule from dye-doped PDLC to white PDLC. The two PDLC films (dye and white PDLC films) were combined together by using UV bond. For demonstration purpose, we placed a piece of white paper behind the bottom substrate to serve as a diffusive reflector.

The electro-optical responses were studied, when an electric field was applied to the sample, using a digital function generator Ellispo resp. RTGS.

Work principle

Figure 2(a,b) shows the work principle of single dye-doped PDLC films in reflective mode at (a) voltage off (b) at voltage ON state. A piece of white paper behind the bottom substrate was attached to serve as a diffusive reflector, whereas Figure 2(c,d) shows new white-dye double PDLC (further abbreviated as W-DPDLC) film with additional PB layer with a piece of white paper behind the bottom substrate to serve as a diffusive reflector at (c) voltage OFF and (d) voltage ON state.

Generally, black dye-doped PDLC can have higher reflectance than normal PDLC structure, because the normal PDLC structure needs an absorption layer to make "On-Off" state in reflective mode driving instead of white reflector. However, for the dye-doped PDLC, the excellent white state is achieved by the combination of white reflector and the transparency mode of dye PDLC. In other words, the disadvantage of dye-PDLC is black state, whose level is higher than normal PDLC films as shown in Figure 2(a). The working principle of W-DPDLC device is shown in Figure 2(c,d). The new constructed W-DPDLC show darker state at off state due to the decrease of the total reflectance made by white PDLC with paper reflector [Fig. 2(c)]. The additional PB layer and white reflective layer act as an active reflective layer. Hence, the total reflectance of W-DPDLC changes when electric field is applied to white-dye PDLC layer. Although it is clear that reflectance of the white reflector remains constant, whether electric field is ON or OFF. Here, the total reflectance is the sum of the combinational structure of W-DPDLC and white reflector. Thus, the "off" (black) state is darker than general dye PDLC film with reflector, and ON state shows more reflectance than general dye PDLC film.

RESULTS AND DISCUSSION

To understand the work of white reflector film as an active reflector, the active reflector structure of pure PDLC of PN393 and TL203 LC with white reflector is shown in Figure 3. To measure the reflectance, the detector was set at 0° (surface normal) while the

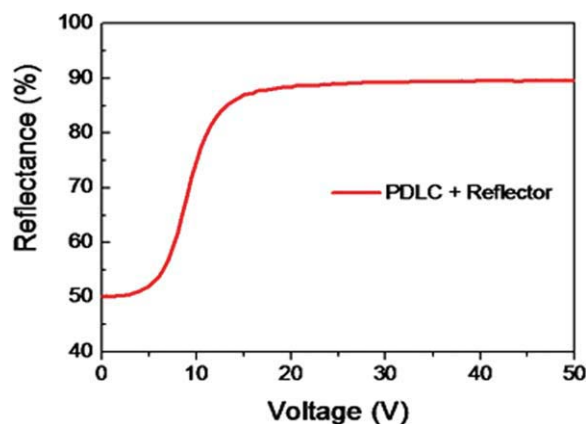


Figure 3 The reflectance of active reflector structure. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

incident parallel white light was moved from 0° to 30° . Figure 3 shows the reflectance of active reflector structure with pure PDLC as a function of applied electric field. Without applied bias, the PDLC show high scattering state. At this scattering state, the use of white reflector makes a reflectance about 50%. As electric field is applied to the active reflector structure, the reflectance increases above 85% due to the transparency of PDLC. This is due to that most light can pass through the PDLC and reflect on through the reflectance layer without loss.

To enhance the optical properties of dye PDLC, we constructed a double-layer white-dye PDLC coupling with the white reflector that works as an active reflector. The schematic view of our constructed assembly is shown in Figure 2. In this, we assembled a white PDLC layer accompanied with white reflector on dye-doped PDLC layer. To compare the results for the reflectance of the single dye-doped PDLC (DPDLC) and double-layer white-dye PDLC (W-DPDLC) are shown in Figures 4 and 5, respectively. The cell thicknesses for the single-layer DPDLC and double-layer W-DPDLC ($20\ \mu\text{m}$ DPDLC + $10\ \mu\text{m}$ W-DPDLC) were set as $30\ \mu\text{m}$, respectively. Figure 4 shows that, when the incident light was at 30° , the single-layer DPDLC at 0.4% dye concentration and at $30\text{-}\mu\text{m}$ cell thickness achieved a reflectivity of 14.53% in its on state at 50 V and 2.9% in the off state. At this angle, the CR was 4.9, which was defined as (reflectivity at white state)/(reflectivity at black state). Compared to single layer, the double-layer W-DPDLC with 0.2% of dye contents and total cell thickness $30\ \mu\text{m}$ ($20\ \mu\text{m}$ DPDLC and $10\text{-}\mu\text{m}$ white-DPDLC) shows the 31.36% reflectivity at on state and 8.2 CR (Fig. 5), which did not use the double W-DPDLC layer, the CR is approximately doubled. The additional white-PDLC layer and white reflector work together as an active reflective layer on voltage ON state. Because of this, the W-DPDLC shows high reflectivity at ON state.

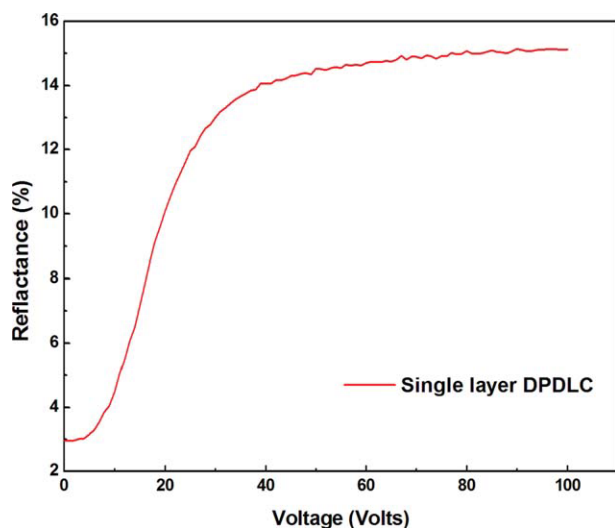


Figure 4 Single layer dye PDLC with 30- μm total cell thickness. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

Because of the active reflector working principle with the double-layer W-DPDL structure, the double PDLC structure shows a good optical property. However, the dichroic dye diffusion happens to the bottom white PDLC structure as the time passes. The diffusion of dye mixing state to the bottom PDLC makes a harmful effect to the role of active reflector and makes nonuniform unstable state finally (inside Fig. 5). Besides this, double-layer W-DPDL film has high-switching voltage of 40 V. Therefore, we need a barrier layer to avoid this situation.

To avoid the diffusion of material from DPDL, we fabricated a new polymer-based barrier device. In this case, we constructed a polymer-based barrier

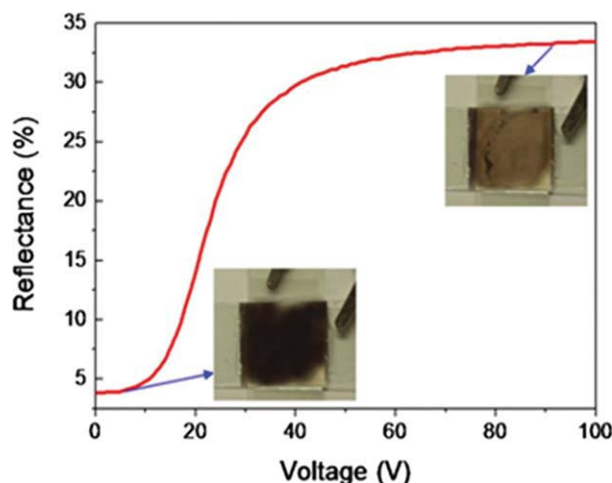
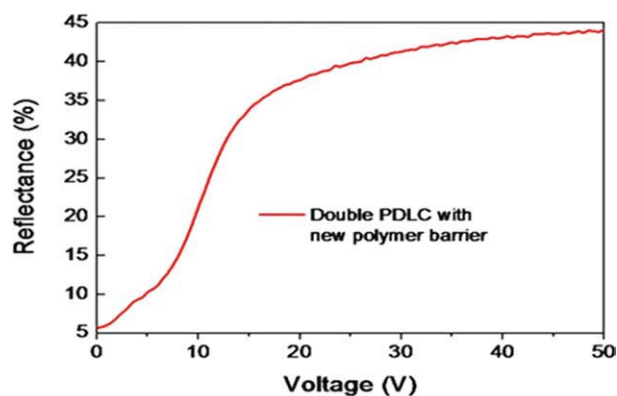
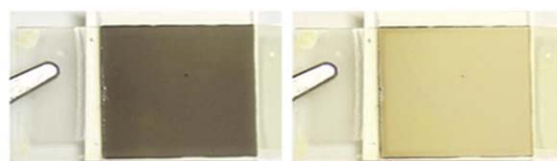


Figure 5 Reflectance of double layer W-DPDL without barrier layer, showing diffusion of dye at zero and at high-applied electric field. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]



(a)



(b)

Figure 6 (a) Double-layer W-DPDL with new polymer barrier (PB) layer and optimal dye ratio; (b) double-layer W-DPDL with PB layer at electric field off (left) and electric field on (right) states. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

layer in-between the two white-dye PDLC films by a regular spin-coating process (stated before). For the purpose, a highly viscous polymer is mixed with IPA of 1 : 1 ratio. This mixture was further coated on white PDLC layer with 500 rpm, with $\sim 2\text{--}3\text{-}\mu\text{m}$ thickness. With the addition of a new very thin PB layer over white-DPDL film, we could stop the diffusion of dye and achieve the high reflectivity and CR. The data for 0.15% dye contents with PB layer on W-D PDLC with total film thickness 30 μm (20 μm dye PDLC and 10- μm white PDLC) is shown in Figure 6(a) and nondiffusion effect of dye on the device with electric field off and on state is shown in Figure 6(b). The introduction of barrier PB layer into double W-DPDL film eliminated the problem of dye diffusion. Moreover, compared to double-layer PDLC films without a barrier layer, it shows the 44% reflectivity and 8.8 CR at comparatively low voltage (>25 V). New material has high viscosity, so that we can make a good and thin barrier layer using simple spin-coating method. The optimization of dye ratio and the high-quality barrier film induces the better result. The process can be applied easily to 5 in. (diagonal) size display device, and we achieved about 90% success yield from 50 samples using research base equipment.

Hence the achievement of double-layer PDLC with combination of white and dye-PDLC films with an additional barrier polymer (PB) films opens a new area of research in the enhancement of CR

of dye-doped PDLC films for various display applications.

SUMMARY AND CONCLUSIONS

In summary, we assembled additional PDLC layer into dye-doped PDLC film with a PB layer as W-DPDL to enhance the CR and reflectivity for dye-doped PDLC for various potential display applications. For our developed structure, we used an additional PB layer between the white and dye-PDLC to stop the diffusion of dye into white PDLC layer. In our developed PB layer white-Dye PDLC structure, we could achieve almost the double CR and high reflectivity at voltage ON state. The main reason is that the combination of white reflector layer and white PDLC work together as an active reflector, which changes its reflectivity with applied electric field. This work can be very useful for the further development of dye PDLC display technology.

The current work was supported by a research grant from Konkuk University, Seoul, Korea in 2011.

References

1. Ferguson, J. L. Dig Tech Pap Soc Inf Symp Disp Int Symp 1985, 16, 68.
2. Drzaic, P. S. J Appl Phys 1986, 60, 2142.
3. Doane, J. W. Liquid Crystals: Applications and Uses, 1; World Scientific: Singapore, 1990; p 361.
4. Kitzerow, H. S. Liq Cryst 1994, 16, 1.
5. Coates, D. J Mater Chem 1995, 5, 2063.
6. Young, J. J.; Bingzhu, Y.; Rhee, J. T.; David, L. C.; Jamil, M. Macromol Theory Simul 2007, 16, 643.
7. Lee, S.-H.; Lim, T.-K.; Shin, S.-T.; Park, K.-S. Jpn J Appl Phys 2002, 41, 208.
8. Mormile, P.; Musto, P.; Petti, L.; Ragosta, G.; Villano, P. Appl Phys B 2000, 70, 249.
9. Hongliu, D.; Zhao, T.; Cheng, Y.; Yuehong, P.; Xu, H.; Shi, G.; Jin, L. Sci China Ser: Chem 2007, 50, 358.
10. Farzana, A.; Jamil, M.; Young, J. J.; Lee, J. W.; Jung, J. E.; Jang, J. E.; Lee G. H.; Park Jin. Sang. J Appl Polym Sci 2011, 121, 1424.
11. Crawford, G. P.; Doane, J. W.; Zumer, S. Handbook of Liquid Crystal Research; Oxford University Press: New York, 1997; p 347.
12. Doane, J. W.; Vaz, N.; Wu, B. G.; Zumer, S. Appl Phys Lett 1986, 48, 269.
13. Im, S. J.; Jin, Y. W.; Sung, J. H.; Park, W. Y. Synth Metals 1995, 71, 2203.
14. Wang, M.; Weidong, L.; Yingfang, Z.; Caiyuan, P. J Phys D Appl Phys 1997, 30, 1815.
15. Yang, K.-J.; Lee, S.-C.; Choi, B.-D. Jpn J Appl Phys 2010, 49, 05EA05.
16. Lin, Y. H.; Yang, J. M.; Lin, Y. R.; Jeng, S. C.; Liao, C. C. Opt Exp 2008, 16, 1777.
17. Bruyneel, F.; Smet, H. De.; Vanfleteren, J.; Calster, A. Van. Liquid Cryst 2001, 28, 1245.
18. Scherschener, E.; Dalchiele, E. A.; Frins, E. M.; Perciante, C. D.; Ferrari, J. A. J Appl Phys 2007, 102, 014502.
19. Lin, Y. H.; Ren, H.; Wu, S. T. Appl Phys Lett 2004, 84, 4083.
20. Cacharelis, Ph.; Frazee, J.; Moore, P.; Luttrell, R.; Flack, R. Solid-State device research conference. Proceeding of the 27th European, Stuttgart, Germany, 1997; p 596-599.
21. Lee, G. H.; Hwang, K. Y.; Jang, J. E.; Jin, Y. W.; Lee, S. Y.; Jung, J. E. Opt Lett 2011, 36, 754.
22. Yang, K.-J.; Kim, K.-P.; Kim, D.-H.; Choi, B.-D. Mol Cryst Liq Cryst 2009, 498, 83.
23. Masutani, A.; Roberts, T.; Schüller, B.; Hollfelder, N.; Kilickiran, P.; Nelles, G.; Yasuda, A.; Sakaigawa, A. Appl Phys Lett 2006, 89, 183514.