Electroluminescent devices with function of electro-optic shutter

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Abstract: The polymer-dispersed liquid crystal (PDLC) was used as a dielectric layer of electroluminescent (EL) device to provide multi-function of electroluminescence and electro-optic shutter. A 50 μm-thick PDLC layer was formed between a transparent electrode and a ZnS:Cu phosphor layer. The electro-optic properties of the EL device were not distorted by the introduction of the PDLC layer. The extraction efficiency of luminescence was improved by more than 14% by PDLC layer. The transmittance of the PDLC was also founded not to be degraded significantly by excitation frequency. Therefore, the electroluminescence of the device was ignited by excitation frequency at a given voltage for full transparency of the PDLC. This device has great potential for applications in transparent displays with the function of a privacy window.

OCIS codes: (230.0230) Optical devices; (230.2090) Electro-optical devices; (160.3710) Liquid crystals; (260.3800) Luminescence.

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#172805 - $15.00 USD Received 18 Jul 2012; revised 24 Aug 2012; accepted 24 Aug 2012; published 29 Aug 2012

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1. Introduction

Luminescence devices, including organic light-emitting diodes (OLEDs) [1], electroluminescent (EL) displays [2, 3] and plasma display panels (PDPs) [4, 5] are considered as suitable technology for transparent displays, because colors are produced by the luminescence of each pixel, and the see-through quality can be modified with the transparent area of the pixel. In contrast, the see-through function is a reason for the drop off of readability of displays and is a limiting factor in transparent displays composed of luminescence devices in their use as smart windows because the inside of the device is hard to blind. To solve these problems, additional panels for optic shutters [6] are required. However, such panels are detrimental to the transparent quality of the display and significantly increase the manufacturing costs. Despite intensive efforts, an ideal method that can lead to cost effectiveness and good quality with high transparency has not been developed. In terms of structure, the merging of electro-optic shutters and display devices such as EL devices is desirable. In this work we propose an EL device with a birefringence-dielectric layer of the polymer-dispersed liquid crystal (PDLC) in order to address this issue. A PDLC layer [7] was formed between a transparent electrode and a phosphor layer; this PDLC layer accumulated charges for the EL and worked as an electro-optic shutter at the same time. We explored the relevance of the PDLC as dielectric layer of EL device. The wavelength and the color coordination of luminescence were shifted by the PDLC in a negligible range. The transmittance of the PDLC was also found not to be degraded at a frequency of around 1 kHz. Therefore, electroluminescence was ignited by increasing excitation frequency at a given voltage for full transparency of the electro-optic shutter by optimizing the PDLC structure. The present idea was motivated by our previous work [8], in which flexible EL devices without oxide dielectric layers were prepared by using UV-curable polymer binder. In the device proposed in paper, charges for electroluminescence were accumulated at a very thin polymer-dielectric layer that was naturally formed between the phosphor and the electrode. This result showed us that the efficiency of electroluminescence could be improved by stacking UV-curable dielectric layers between the phosphor layer and the electrode. This novel structure was extended to an EL device with an electro-optic shutter using birefringence materials of PDLCs as a dielectric layer. We designated these structures as switchable birefringence-dielectric electroluminescent (SBDEL) devices.

2. Concept of SBDEL devices

The conceptual structure of the suggested SBDEL device is schematically depicted in Fig. 1. Instead of a conventional dielectric oxide layer [9] [Fig. 1(a)], a switchable birefringence-dielectric material of PDLC layer is formed on the phosphor layer [Fig. 1(b)]. Then the PDLC layer works as a dielectric electrode for charge accumulation and as an electro-optic shutter at the same time.

#172805 - $15.00 USD Received 18 Jul 2012; revised 24 Aug 2012; accepted 24 Aug 2012; published 29 Aug 2012

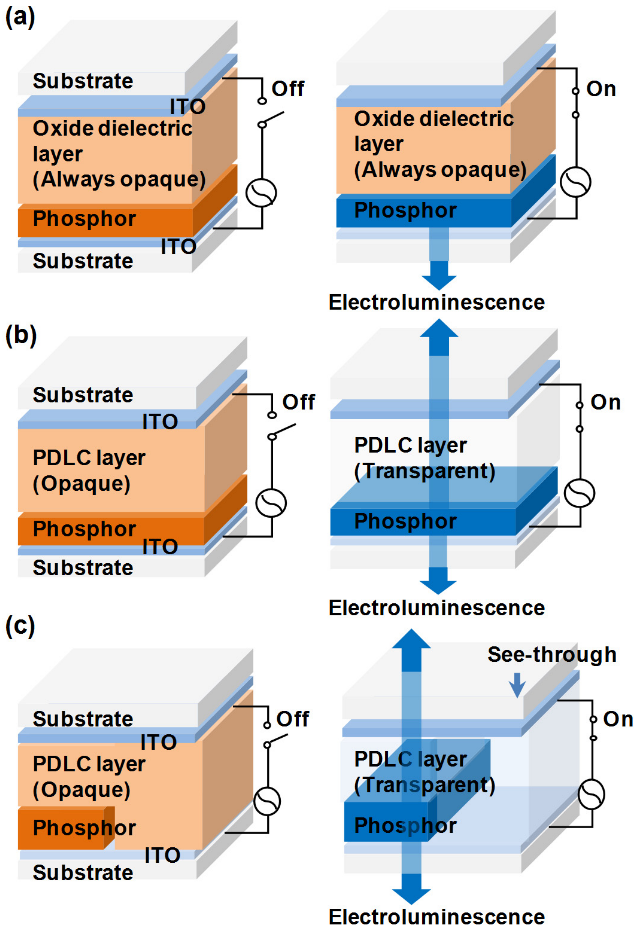

Fig. 1. (a) Conventional EL device (off and on state) using oxide composite as dielectric layer. (b) SBDEL device (off and on state) using birefringence composite of PDLC instead of oxide composite as dielectric layer. (c) Pixel design of SBDEL device (off and on state) for see-through.

In the conventional structure, a dielectric layer composed of an oxide composite is always opaque; therefore, it is difficult to produce a function of transparency with a privacy window. On the contrary, in the proposed structure, a milky PDLC layer can switch to the transparent state at the voltage applied for electroluminescence; then, the device can pass light through the dielectric layer of PDLC, as shown in Fig. 1(b). Consequently, a see-through function can be provided to the device. In particular, a smart window using a luminescence device with a privacy window can be achieved by proper design of the luminescence pixels [Fig. 1(c)]. The fabrication process is also expected to be cost effective and damage free for EL devices because the PDLC layer can be formed by photopolymerization-induced phase separation (PIPS) method, which is a rather simple and room temperature process [10].

3. Experimental

We printed a 100 µm-thick blue-emitting ZnS:Cu phosphor [11] patterns with dimensions of 5 x 40 mm^2 on a glass substrate (50 x 50 mm^2), with surfaces coated with an indium-tin-oxide (ITO) layer by a conventional screen-printing method. The printed phosphor pattern was dried at 120 °C for 10 min in an oven to remove the organic components. Then, UV (365 nm, 200 mW/cm^2) curing for 120 s at room temperature was followed by coating of the PDLC mixture [12] and laminating of the PET film with an ITO layer on surface. We prepared two samples with stripes and 'DGIST' logo pattern. The target thicknesses of PDLC layer were 150 and 50 μm, respectively. We also prepared PDLC cells with a 7 μm cell-gap by vacuum filling method described elsewhere [13] to measure the transmittance a function of voltage in the frequency range of 100 Hz to 50 MHz.

The phosphor paste suitable for the printing process was prepared by blending of 70 g of ZnS:Cu phosphor (Osram Sylvania) and 30 g of a mixture of α-terpineol (5 wt% KANTO Chemical Industries, Co. Inc.) and ethyl cellulose (95 wt% Sigma Aldrich, Co.). The PDLC mixture was prepared by blending a nematic LC (E7, Merck, $\Delta n = 0.225$, $\Delta\varepsilon = 14.3$) and the UV-curable prepolymer PN393 (Merck) at 80:20 wt% ratio.

The emission spectra of devices were evaluated by spectrometer (CS-1000, Canon). The cross-sectional images of devices were investigated by optical microscopy (OM, Nikon LV-100) and scanning electron microscopy (SEM, Hitachi SU8020). The transmittance of PDLC was measured with a photodiode after placing PDLC cells normal to the direction of collimated beam of He = Ne laser (632.8 nm). The drive signal and the response of the photodiode were monitored with a digital storage oscilloscope (Hitachi VC-6023).

The effects of PDLC layer on electro-optical property were directly compared by measuring emission spectra at position P1 and P2 as shown in Fig. 3(a). The electroluminescence generated at the phosphor layer passes through the PDLC layer and the transparent substrate with the ITO electrode on the surface to reach the front side, P1. On the reverse side, the electroluminescence to reach the back side, P2 passes through the transparent substrate with the ITO electrode on the surface. Therefore, we can directly compare the emission spectra measured at the two positions of P1 and P2 in order to identify the influence of the PDLC layer.

4. Results and discussions

4-1. Optical properties of SBDEL devices

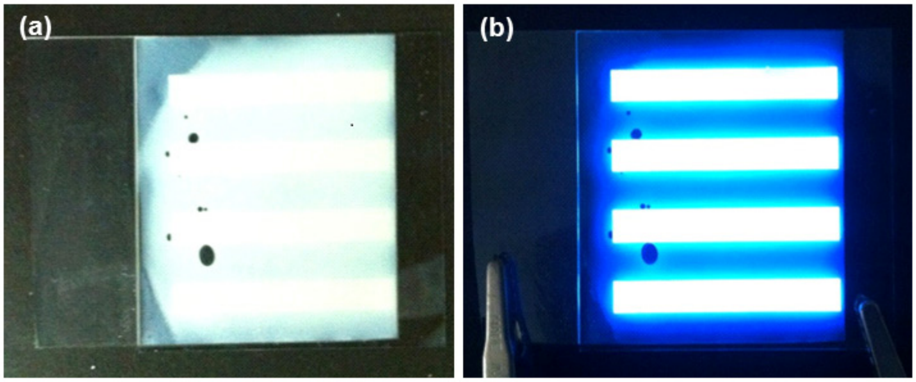

Fig. 2. (a) Photographs of the prepared SBDEL device as-prepared. (b) The blue luminescence at 150 V and 1 kHz from the corresponding device

The prepared SBDEL device is shown in Fig. 2(a); this device successfully emitted blue luminescence [Fig. 2(b)] under optimized condition of 150 V and 1 kHz. Figure 3(b) shows the intensity of the luminescence as a function of the applied voltage at various frequencies. The luminescence began to appear at the applied voltage of 50 V, which voltage value did not critically change with the variation of the excitation frequency. The intensity of the electroluminescence, however, was drastically increased with the rise in excitation frequency and applied voltage. This behavior is nearly the same as the behavior of conventional EL devices using oxide dielectric layer [14].

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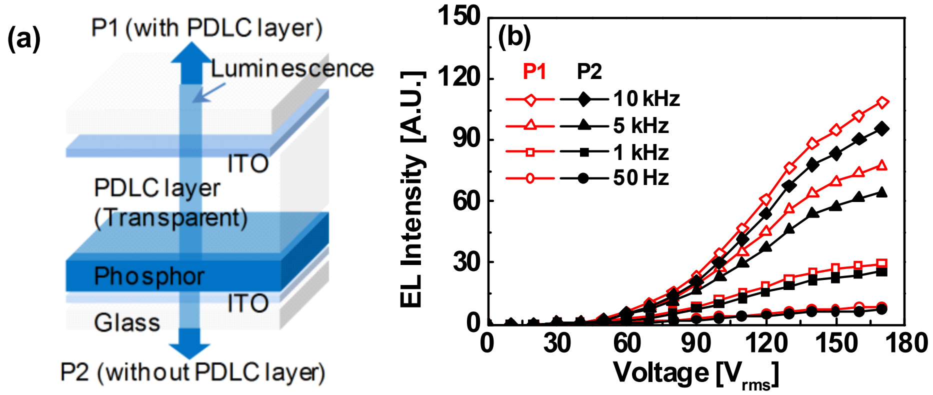

Fig. 3. (a) Schematic illustration of the measurement of electro-optic property of SBDEL device: P1(with PDLC layer), P2 (without PDLC layer) (b) Intensity of emission spectra as a function of applied voltage at various frequencies (50 Hz to 10 kHz).

Besides, we investigated the influence of the PDLC layer on the emission spectra because the electro-optical property of the device can be changed with the birefringence refractive index [15].

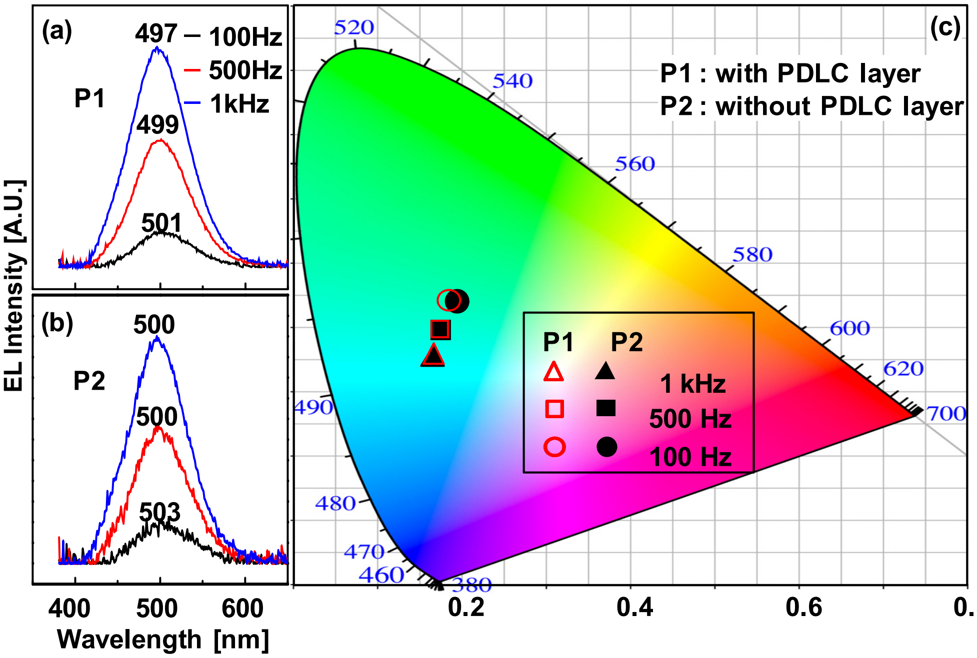

Fig. 4. (a) The emission spectra measured at the top position, P1 (with PDLC layer) at various frequencies (150 V and 100 Hz, 500 Hz, 1 kHz). (b) The emission spectra measured at the bottom position, P2 (without PDLC layer) at various frequencies (150 V and 100 Hz, 500 Hz, 1 kHz). (c) Color coordinates (CIE 1931 color space) measured at P1 (150 V and 100 Hz (○), 500 Hz (□), 1 kHz (△)) and P2 (150 V and 100 Hz (●), 500 Hz (■), 1 kHz (▲)).

Figures. 4(a) and 4(b) show the emission spectra of the device measured by spectrometer in a frequency range of 100 Hz to 1 kHz at P1 and at P2, respectively. Regardless of the presence of a PDLC layer, the emission spectra exhibited similar spectral shapes and the intensity increased systematically with increasing frequency. It was also observed that the

#172805 - $15.00 USD Received 18 Jul 2012; revised 24 Aug 2012; accepted 24 Aug 2012; published 29 Aug 2012

center peak of the luminescence was gradually blue-shifted, with an increasing excitation frequency from 501 to 497 nm at P1 [Fig. 4(a)], and from 503 to 500 nm at P2 [Fig. 4(b)]. Such a behavior of the blue-shift is ascribed to the emission property of ZnS:Cu phosphor, which has two defects in terms of emission, one for green and the other for blue [16]. The blue emissions became more intense due to the increasing excitation frequency [17]. On the other hand, the difference of center peak by the PDLC layer measured at 1 kHz was identified about 3 nm, this value was considered small enough not to induce considerably color distortion of luminescence. In fact, the change of the color coordinates of the emission spectra measured at P1 and P2 as given by the Commission Internationale de l'Eclairage (CIE) 1931 color space was negligible; these coordinates were (0.177, 0.440) and (0.188, 0.443) at 100 Hz, (0.165, 0.396) and (0.167, 0.391) at 500 Hz, and (0.160, 0.361) and (0.160, 0.354) at 1 kHz shown in Fig. 4(c). As a result, it is known that the electro-optic properties of the EL device are not seriously distorted by the introduction of the PDLC layer.

4-2. Extraction efficiency by the PDLC layer

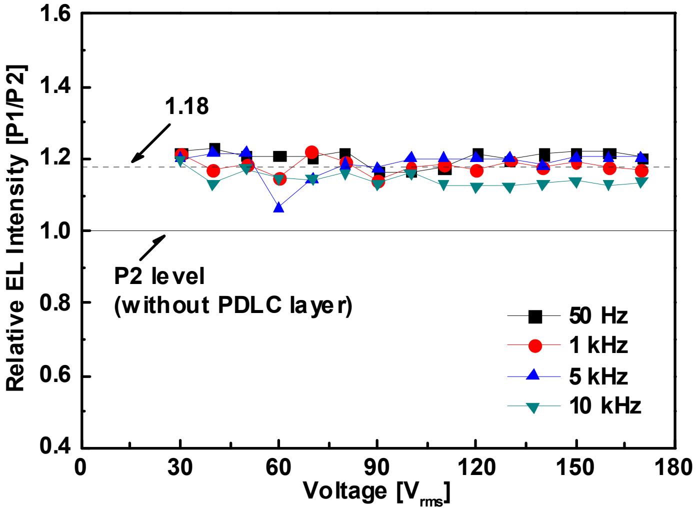

Fig. 5. Relative intensity of emission spectra measured at P1 (with PDLC layer) and P2 (without PDLC layer) as a function of excitation voltage at various frequencies.

Above mentioned Fig. 3(b) shows that the intensity of P1 (with PDLC layer) rapidly increased, compared to that of P2 (without PDLC layer). The emission intensities measured at P1 and P2 are compared in Fig. 5 to investigate the increasing rate as a function of the excitation voltage at various frequencies. The relative intensity of P1/P2 was evaluated as 1.18 in an average value, and revealed similar value with regardless of excitation voltage. This tendency was repeated at different frequencies at 50 Hz to 10 kHz, which indicated that PDLC layer improves extraction efficiency of electroluminescence. This result could be caused not by internal factors but by structural factors such as the interface between the PDLC and the phosphor since the intensities are measured at different sides with the same device. The optical property of the top and bottom substrates also could be a reason because the measured transmittances of PET and glass are 92% and 88% around 500 nm. Considering this effect it can be concluded that the extraction efficiency by PDLC layer was improved by more than 14%.

A polymer usually forms a continuous interface with inorganic materials. In this device polymer can form a continuous interface with phosphor; as a result, the centers for light scattering are considered to decrease at the interface between the PDLC and the phosphor layer.

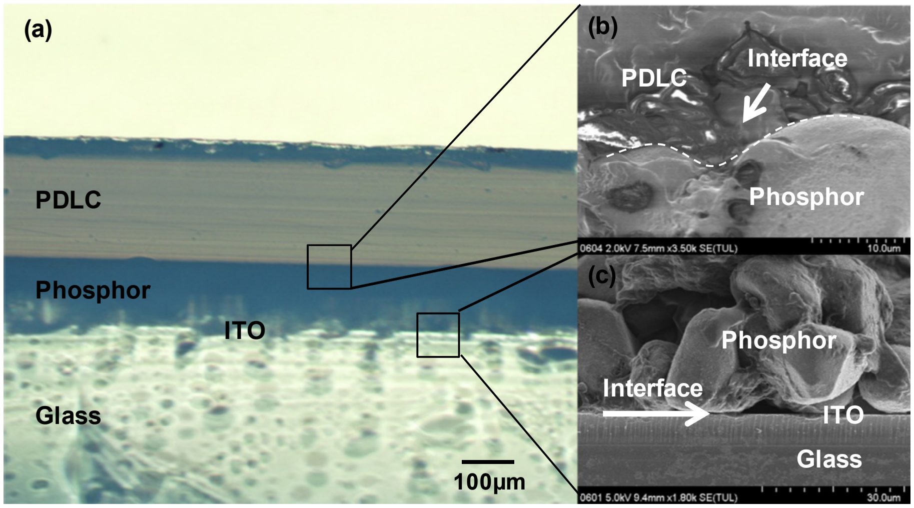

Fig. 6. (a) Optical microscope image of whole device structure. (b) Magnified SEM image of the interface between the PDLC layer and the phosphor (white dashed line). (c) Magnified SEM image of the interface between the phosphor and the ITO (white arrow).

For a detailed analysis, we investigated the cross-sections of the device by using SEM and OM. Figures 6(a) and 6(b) provide the interface images between the PDLC and the phosphor layer, which is continuously formed. On the contrary, the interface between the phosphor and the transparent electrode of the ITO layer consists of a porous structure and air gaps [Fig. 6(c)], which structures were identified as inevitable defects because they were formed by residual gases generated during the thermal process for formation of the phosphor film [18] and by the shape of the phosphor particles, respectively. The pore and air gap at the interface is recognized as a barrier to carrier injection into the phosphor layer [19] and then as a scattering center for luminescence. Since the PDLC layer was formed on the phosphor by no residual gas process of UV curing at room temperature, and because this PDLC layer made contact with the phosphor in a liquid state, interfaces with continuous structures were formed, which is an effective way to decrease not only the injection barrier but also the scattering center by reducing the air gap at the interface.

4-3. Multi-function of SBDEL devices

The outstanding feature of the SBDEL device is that a PDLC layer can operate under a condition different from electroluminescence. PDLC operates in a low voltage range but electroluminescence is excited at high voltage and frequency. This feature enables a smart window made of a SBDEL device to perform two functions of privacy window and electroluminescence device such as display in a separated state by optimizing the driving conditions. In the prepared device [Fig. 2], however, we were not able to observe such an independent operation of electroluminescence with the electro-optic shutter at the applied voltages, which result was contradictory to our anticipation. There might be several reasons for this problem. We concluded from the OM image shown in Fig. 6(a) that the problems were not caused by faults in the device, such as electrical shorts, disconnection at interfaces, and so on, because the thickness of the PDLC, measured over 150 µm, was thick enough to prevent electrical breakdown. Conversely, it is known that the PDLC layer was too thick to work at an applied voltage lower than the excitation voltage of electroluminescence. Therefore, a high voltage that far exceeds the excitation voltage for electroluminescence will be needed for full transparency of the PDLC.

On the other hand, these conclusions suggest that it will be possible to prepare a novel device with multi-functions of electro-optic shutter and electroluminescence through a precise design of the constituents. In order to meet the necessary conditions we needed to know details about the properties of the PDLC in the range of the applied voltage and frequency.

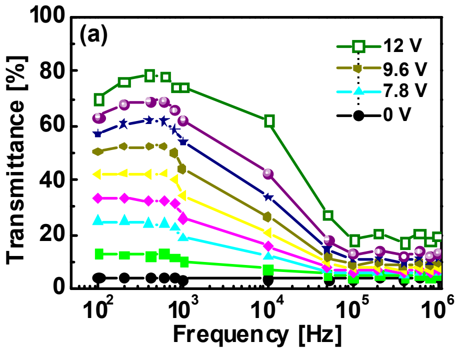

Fig. 7. (a) Transmittance-frequency (T-F) curve of the PDLC at a variety of voltage. (b) Transmittance-voltage curve of the PDLC at a variety of frequency. (Thickness of PDLC: 7 μm)

Figure 7(a) shows the results of the transmittance-frequency (T-F) curve of the PDLC cell at various voltages. It is known that the transmittance is drastically decreased in the high frequency range over 100 kHz. This result is considered to stem from the change of the refractive index of LC in the PDLC due to the increasing frequency. The refractive index of LC depends on the dielectric constant, which changes with the frequency [20]. However, the transmittance was not seriously degraded in a frequency range below 1 kHz. The transmittance-voltage (T-V) curve [Fig. 7(b)] for a variety of frequencies also supported this result, and convinced us that the electroluminescence can be independently controlled with the PDLC by optimizing the thickness of the PDLC to make it fully transparent at a voltage over V_{th} for electroluminescence. This is because, at a voltage over V_{th}, the intensity of the electroluminescence strongly depends on the excitation frequency, as shown in Fig. 3(b).

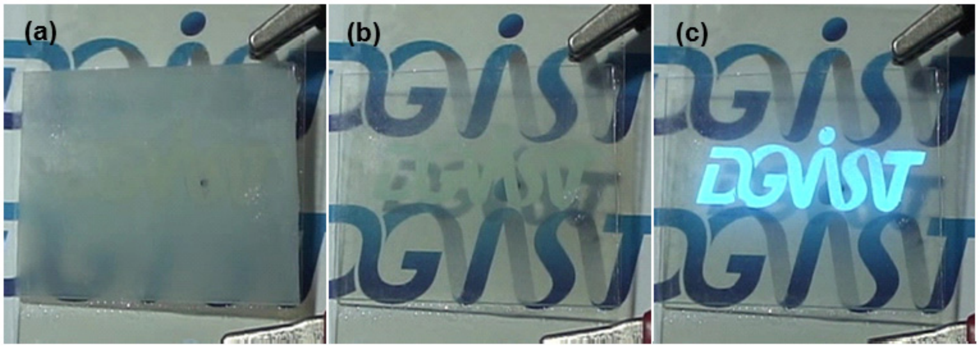

Fig. 8. (a) SBDEL device with a phosphor pattern of ‘DGIST’ blinds the see-through image when the voltage is not applied. (b) The device showing a see-through image when the applied voltage is increased to 150 V at 100 Hz. (c) The electroluminescence of the phosphor pattern of ‘DGIST’ was dramatically enhanced by increasing the excitation frequency from 100 Hz to 1 kHz at 150 V.

The driving voltage of the PDLC layer depends on the film thickness [21]. The driving voltage for full transparency by unit thickness was calculated at 1.7 V/μm. This meant that a PDLC layer with a thickness lower than 80 μm should be formed in order for the PDLC to be fully operated below 150 V.

#172805 - $15.00 USD Received 18 Jul 2012; revised 24 Aug 2012; accepted 24 Aug 2012; published 29 Aug 2012

To test our concept we coated a 50 μm-thick PDLC layer on an ITO substrate with a phosphor pattern of the logo ‘DGIST’. The PDLC mixture was directly contacted onto ITO surface, which had no phosphor pattern, to test the see-through function.

Photographs of the device are shown in Fig. 8. The PDLC was milky when the voltage was not applied [Fig. 8(a)] and became completely transparent at conditions of 150 V and 100 Hz [Fig. 8(b)]. In this state, we increased the frequency up to 1 kHz to enhance the electroluminescence; at this point the intensity dramatically increased, as shown in Fig. 8(c). If we use reverse modes PDLC, it is also possible to switch the PDLC layer from a state of transparency to a milky state, as has been reported elsewhere [22]. In addition, new driving scheme such as a two-frequency addressing mode to control the transparency of the PDLC can be considered for expanding the possible range of the device applications.

5. Conclusions

We proposed a novel device with a multi-function of electric blind and electroluminescence by using PDLC as a dielectric layer of an EL device; this device can be used in transparent displays to provide the function of a privacy window. The difference of center peak and color of luminescence due to the PDLC layer were found to be negligible. The extraction efficiency of the luminescence was improved by more than 14% by the PDLC layer, because the polymer in PDLC formed a continuous interface with the phosphor. With a precise design of the PDLC layer, it was possible to independently control the electroluminescence with the function of the electro-optic shutter. We believe that these kinds of multi-functional devices have strong potential for use in transparent displays with the function of a privacy window.

Acknowledgments

The authors would like to thank Mr. K. J. Yang for the preparation and property measurement of the PDLC cells. This work was supported by the basic research program (11-NB-02) through the Daegu Gyeongbuk Institute of Science and Technology (DGIST), funded by the Ministry of Education, Science and Technology (MEST) of Korea.