Suppression of the color shift of microcavity organic light-emitting diodes through the introduction of a circular polarizer with a nanoporous polymer film

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ABSTRACT
A circular polarizer (CP) film modified by a nanoporous polymer film (NPF) was applied to improve the viewing angle characteristics of a top-emission organic light-emitting diode (TEOLED) without any serious pixel blur phenomenon. From this approach, the angular color shift (Δu′v′) was significantly improved from 0.096 to 0.056, and the angular luminance distribution was also expanded to the Lambertian distribution direction. The black color had a slightly faded tint, however, like the turn-off condition of the plasma display panel (PDP), regardless of the introduction of NPF below or above the CP. Very interestingly, the surface reflectance of the device with NPF-coated CP was not significantly different from that of the device with non-NPF CP. The pixel blur level according to the distance between the scattering medium and the light source was also investigated. As a result, it was found that the pixel blur phenomenon can be greatly suppressed by applying a thinner encapsulation lid with an NPF film.

1. Introduction
Organic light-emitting diodes (OLEDs) have been attracting much attention for display and lighting applications. In addition, they may be applicable to various fields due to their numerous advantages, such as self-emission, low power consumption, high contrast ratios, fast response time, thinness, and overall flexibility [1–6]. The external quantum efficiency (EQE) of OLEDs, however, cannot be typically greater than 20% because of the significant level of light loss originating from the surface plasmon coupling mode, substrate mode, waveguide mode, absorption mode, etc. [7–9]. Hence, many groups have exerted much effort to improve the device efficiency by ruling out these problems. This has resulted in highly efficient OLED devices showing extremely high EQEs. Nevertheless, the major panel makers are still reluctant to use such technologies even though some outcoupling approaches using microlens arrays (MLAs), high-index glasses, and corrugated substrates can easily increase the EQEs [10–20]. A typical reason for this is that using these techniques causes a serious pixel blurring effect. Therefore, most companies are making microcavity devices to increase the efficiency. In fact, this technology is inevitably used because of the unavoidable top-emitting structure due to a complicated pixel circuit. Unfortunately, although the microcavity device can improve the efficiency, it has a problem: the viewing angle dependency is deteriorated due to its strong resonator behavior [21,22]. To solve this problem, it is necessary to develop a technology for widely spreading the direction of light. In particular, it is desirable to add this technology as a component. As such, the authors have developed a technology for improving the existing circular polarizer (CP) film. Reported herein is the circular polarizing film, in which a nanoporous polymer film (NPF) is integrated to suppress the device’s viewing angle dependence.

2. Experiment
2.1. NPF preparation
Cellulose acetate butyrate (CAB) (butyryl content: 35–39 wt. %; acetyl content: 16–19 wt. %; Mw: 16,000 g mol⁻¹) was purchased from ACROS and used for the preparation of a scattering layer. CAB solution was prepared in chloroform with an 8 wt. % concentration. The NPF was prepared through the simple spin coating process, under a humid atmosphere and with a continuous supply of water droplets, as reported in previous studies [23–28]. Meanwhile, the NPF was dried for over 10 min at room
temperature after the spin coating process. As a result, a 2-μm-thick NPF was obtained on the glass substrate, and it showed an optical haze value of about 40%. Especially, the pores in the NPF were randomly distributed in the CAB base film with an about 300–500 nm diameter, as shown in Figure 1(a,b). The morphology and pore distribution of the polymer films were investigated using the field emission scanning electron microscopy (FESEM, S-4700, Hitachi) and atomic force microscopy (AFM, XE-100, Park System) technologies. The haze of the porous polymer films on the bare glass was measured using a haze meter (NDH-5000, Nippon Denshoku Industries).

Table 1. Optical characteristics of the devices in this study.

| Items                        | Reference | Device A | Device B | Device C |
|------------------------------|-----------|----------|----------|----------|
| Operating voltage$^a$ (V)    | 3.9       | 4.5      | 4.8      | 4.8      |
| Current efficiency$^a$ (cd A$^{-1}$) | 47.1     | 23.0     | 17.7     | 16.8     |
| Power efficiency$^a$ (lm W$^{-1}$) | 26.6     | 12.0     | 9.9      | 9.7      |
| $\Delta u'$ $^{a,b}$         | –         | 0.0094   | 0.057    | 0.056    |

$^a$Measured at 1000 cd m$^{-2}$.

$^b$CIE 1976 color space.

2.2. OLED fabrication

Clean glass pre-coated with ITO/Ag/ITO anode layers was used as a substrate. Line patterns of the anode materials were formed on the glass using photolithography. A bank layer was also formed on the anode and the glass substrate using photolithography, to define the pixel aperture area using photoresist. The glass substrate, anode, and bank layer were cleaned through sonication in isopropyl alcohol and acetone, rinsed in deionized water, and then irradiated in a UV-ozone chamber. The organic materials were thermally deposited in a vacuum chamber under around 5 $\times$ 10$^{-7}$ Torr pressure, and their deposition rate was in the 0.5–1 Å s$^{-1}$ range. Liq as an EIL and Mg and Ag for the cathode were deposited at the 0.15, 1.8, and 0.2 Å s$^{-1}$ rates under around 10$^{-6}$ Torr, respectively. The current density–voltage (J–V) and luminance–voltage (L–V) data of the OLEDs were measured using a Keithley SMU 238 and a Minolta CS-2000, respectively. The electroluminescence (EL) spectra and the Commission International De’Eclairage (CIE) color coordinates were obtained using a Minolta CS-2000A spectroradiometer. The pixel size of the OLED was 4 mm$^2$ for all the samples that were used in this study.

3. Results and discussion

3.1. Optical simulation

Top-emission organic light-emitting diodes (TEOLEDs) with strong microcavity characteristics were fabricated using two important interferences: wide-angle interference and multiple-beam interference. Wide-angle interference occurs between directly emitted light and reflected light (two-beam interference). It is significantly affected by the distance between the dipole of the emitter and the highly reflective anode. Multiple-beam interference (Fabry–Perot interference), on the other hand, occurs between two reflective electrodes and is caused by the infinite reflection between a semi-transparent electrode and a highly reflective electrode. The theoretical spectrum in free space is expressed as shown below.

$$I_{ext}(\theta, \lambda, z_0) = \frac{T_c}{1 + R_a R_c - 2 \sqrt{R_a R_c \cos(\Delta \varphi_{FP})}} \times (1 + R_a + 2 \sqrt{R_a \cos(\Delta \varphi_{TI})}) \times I_{int}(\theta, \lambda),$$

$$\Delta \varphi_{FP} = \varphi_a + \varphi_c - \sum_{i=1h}^{4\pi n_i d_i \cos \theta} \frac{4\pi n_i d_i \cos \theta}{\lambda},$$

$$\Delta \varphi_{TI} = \varphi_a - \frac{4\pi n_{org} z_0 \cos \theta}{\lambda},$$

where $\Delta \varphi_{FP}$ and $\Delta \varphi_{TI}$ are the phase terms of the Fabry–Perot and two-beam interference, respectively; $\theta$
Figure 2. (a) Relative luminance contour plots of the microcavity TEOLED with a red phosphorescent emitter as functions of the HTL vs. ETL thicknesses obtained through optical simulation. The red solid box designates the optimal thickness range of HTL/ETL to satisfy the second-order condition. (b) Simulated luminance of the red microcavity TEOLED as a function of the HTL thickness.

is the internal observation angle from the normal direction of the microcavity; $z_0$ is the distance between the dipole of the emitter and the highly reflective anode; $T_c$ is the transmittance of the cathode; $R_a$ and $R_c$ are the reflectance values at the anode-organic and cathode-organic interfaces, respectively; $\varphi_a$ and $\varphi_c$ are the phase changes that occur with the reflections at the anode-organic and cathode-organic interfaces, respectively; and $I_{int}(\theta, \lambda)$ is the initial emissions spectrum in the free space at $\lambda$. In Equation (1), the term $\frac{T_c}{1+R_aR_c-2\sqrt{R_aR_c}\cos(\Delta\varphi_{FP})}$ implies Fabry–Perot interference; meanwhile, the term $1+R_a+2\sqrt{R_c}\cos(\Delta\varphi_{TI})$ is the two-beam interference. In Equation (2), $n_i$ and $d_i$ are the refractive index and the thickness of the organic layer, respectively. In Equation (3), $n_{org}$ denotes the refractive index of the organic material. Optical simulations were conducted to investigate the highly efficient microcavity structures for phosphorescent red OLEDs before fabricating the device, as shown in Figure 2(a). In this simulation, the second-order microcavity condition with a thick hole transport layer (HTL) and a thin ETL was chosen to prevent the dust issue caused by low organic thickness. Based on the simulation results, the radiance value could be maximized when the HTL thickness was between 210 and 230 nm and when the ETL thickness was between 40 and 60 nm. From these conditions, a relatively thin (53 nm) ETL was fabricated to adopt the thick (215 nm) HTL, as shown in Figure 2(b).

3.2. Optical performance of NPF

The Reference device with strong microcavity characteristics was prepared. As the driving voltage of the OLED device is normally increased if the distance between the reflective anode and the emitting layer is adjusted with only the HTL material to match the second-order microcavity optical length (215 nm), HATCN, which can function as a charge generator in the HTLs, was deposited, as shown in Figure 3(a,b). To investigate the optical characteristics by applying various conditions, four types of OLED devices were prepared, including the Reference device, as shown below.

**Reference:** NPB (75 nm)/HATCN (7 nm)/NPB (75 nm)/HATCN (7 nm)/NPB (51 nm)/Bebq$_2$:I$_r$(phq)$_2$(acac) (20 nm, 3%)/BPhen (53 nm)/Liq (1 nm)/Mg:Ag (14 nm)/NPB (60 nm)/encapsulation glass (500 μm)

**Device A:** NPB (75 nm)/HATCN (7 nm)/NPB (75 nm)/HATCN (7 nm)/NPB (51 nm)/Bebq$_2$:I$_r$(phq)$_2$(acac) (20 nm, 3%)/BPhen (53 nm)/Liq (1 nm)/Mg:Ag (14 nm)/NPB (60 nm)/encapsulation glass (500 μm)/CP (160 μm)

**Device B:** NPB (75 nm)/HATCN (7 nm)/NPB (75 nm)/HATCN (7 nm)/NPB (51 nm)/Bebq$_2$:I$_r$(phq)$_2$(acac) (20 nm, 3%)/BPhen (53 nm)/Liq (1 nm)/Mg:Ag (14 nm)/NPB (60 nm)/encapsulation glass (500 μm)/NPF (2 μm)/CP (160 μm)

**Device C:** NPB (75 nm)/HATCN (7 nm)/NPB (75 nm)/HATCN (7 nm)/NPB (51 nm)/Bebq$_2$:I$_r$(phq)$_2$(acac) (20 nm, 3%)/BPhen (53 nm)/Liq (1 nm)/Mg:Ag (140 nm)/NPB (60 nm)/encapsulation glass (500 μm)/CP (160 μm)/NPF (2 μm)

Figure 4(a) shows the J–V–L characteristics of the Reference, Device A, Device B, and Device C. Obviously, NPF does not affect the electrical properties because NPF is applied on the encapsulation glass. The driving voltages required to reach 1000 cd m$^{-2}$ were 3.9, 4.5, 4.8, and 4.8 V for Reference, Device A, Device B, and Device C, respectively. The current efficiencies and power efficiencies required to reach 1000 cd m$^{-2}$ were 47.1 cd A$^{-1}$ and...
26.6 lm W$^{-1}$ for Reference, 23.0 cd A$^{-1}$ and 12.0 lm W$^{-1}$ for Device A, 17.7 cd A$^{-1}$ and 9.9 lm W$^{-1}$ for Device B, and 16.5 cd A$^{-1}$ and 9.7 lm W$^{-1}$ for Device C, as shown in Figure 4(b,c). From these results, it was found that the transmittance of CP (e.g. 48%) may significantly reduce the current efficiency when comparing Reference and Device A. Interestingly, the current efficiency of Device B and Device C was reduced by 23.1% and 28.3%, respectively, compared to Device A, while the power efficiency was reduced by 17.5% and 19.2% because NPF slightly causes enlarged angular luminance distribution towards the Lambertian distribution direction, as shown in Figure 4(d). Meanwhile, the difference in the optical characteristic between Device B and Device C is minimal, and this means that the optical characteristics of the devices are not affected by the location of the NPF coating (e.g. below or above the CP). To evaluate the efficiency of suppression of the viewing angle dependence using NPF, the angular EL spectrum from the viewing angle of 0–60° was measured for Device A, B, and C, as shown in Figure 5(a–c), respectively. The angular EL spectrum shifts of Device B and C were significantly improved to 10 and 9 nm while that of Device A (without NPF) was 17 nm. In addition, the angular color shifts of Device B and C [from 0 to 60°, Δu′′v′′ (CIE 1976): 0.057 and 0.056] were much more improved compared to that of Device A [Δu′′v′′ (CIE 1976): 0.094]. From these angular characteristics of the devices, it was found that the proposed NPF has excellent performance in suppressing the viewing angle dependence of strong microcavity OLEDs regardless of the NPF location in the CP.

### 3.3. Evaluation of the pixel blur in the devices

The pixel blur levels of the devices were also examined to determine if the proposed NPF could be applied to the display application. Normally, the conventional light extraction film or diffuser film cannot be applied in OLED displays because it is difficult to distinguish a pixel...
edge if such films are applied. This is because the pixel blur phenomenon prevents the technology from being applied to the display due to the decreased image resolution and readability capability. Thus, it is mandatory to suppress these effects to be able to apply the diffuse film or light extraction film to the display application. To quantitatively analyze the pixel blur level, the relative luminance data were measured with a charge-coupled device (CCD) to compare the luminance distributions of the pixel boundaries. In addition, devices made with the conventional MLAs and that had a regular hemispherical pattern with an 80 μm diameter were prepared for comparison purposes. A thin-film encapsulation with 50 μm PET instead of 500 μm glass was additionally prepared, as shown below.

**Device D**: NPB (75 nm)/HATCN (7 nm)/NPB (75 nm) /HATCN (7 nm)/NPB (51 nm)/Bepq2: Ir(phen)2(acac) (20 nm, 3%)/BPhen (53 nm)/Liq (1 nm)/Mg:Ag (14 nm)/ NPB (60 nm)/encapsulation glass (500 μm)/MLA (180 μm)/CP (160 μm)

**Device E**: NPB (75 nm)/HATCN (7 nm)/NPB (75 nm) /HATCN (7 nm)/NPB (51 nm)/Bepq2: Ir(phen)2(acac) (20 nm, 3%)/BPhen (53 nm)/Liq (1 nm)/Mg:Ag (14 nm)/ NPB (60 nm)/thin-film encapsulation (50 μm)/NPF (2 μm)/CP (160 μm)

When only Reference, Device A, Device B, and Device E were visually observed through picture images, it was found that there was no serious pixel blur phenomenon in Device A, Device B, and Device E compared with Reference, as shown in Figure 6(a–c,e). There was a serious pixel blur phenomenon, however, in Device D, which had an MLA attached beneath the CP, as shown in Figure 6(d). As it was found that the distance between the light source and the scattering layer is the key factor for the pixel blurring effect, it can be concluded that it could be diminished if such distance is very short, as reported in the previous study [28]. Thus, a thin 50 μm polyethylene terephthalate (PET) film was applied as a lid for encapsulation. To prevent degradation by the penetration of H2O or/and O2, a 50 nm Al2O3 layer was additionally deposited on the PET surface through atomic layer deposition (ALD). To quantify the pixel blur level of the devices, the luminance distribution from the lighting pixel images was analyzed using a CCD camera, as shown in Figure 7(a). Furthermore, the distance, which is one-tenth of the luminance of the pixel edge, was measured, as shown in Figure 7(b). In other words, the distances in which the luminance contrast ratio becomes 10:1 from the pixel edge were compared, and they were called ‘pixel blur distances’ to quantitatively compare the pixel blur.

**Figure 5.** (a) Color shift ($\Delta u'v'$ in CIE1976) of the fabricated devices, and peak wavelength shift of (b) Device A, (c) Device B, and (d) Device C with viewing angle variation.

**Figure 6.** Electroluminescent pixel image comparison of the various devices in this study: (a) Reference; (b) Device A; (c) Device B; (d) Device D; and (e) Device E.
levels. Using this approach, Device D showed a 650 μm pixel blur distance, and Device B showed 410 μm. From this result, it was found that NPF is superior to the MLAs in suppressing pixel blur. Moreover, the pixel blur distance of Device E using NPF and thin PET encapsulation was 170 μm, which is much smaller than that of the device using NPF on the glass encapsulation. This pixel blur distance of Device E is minimal compared to those of Reference (100 μm) and Device A (130 μm).

3.4. Investigation of the reflectance of the devices

In general, when the light coming from outside enters through the transparent lid for OLED encapsulation, it can be directly reflected by a reflective material like a metal electrode. Therefore, it is essential to use CP in OLED display application to prevent the dropping of the contrast ratio under bright outdoor conditions. To verify if the contrast ratio gets worse after introducing NPF, the reflectance values of four samples with NPF formed beneath or above the CP were measured, as shown below.

**Sample A**: Al (100 nm)/glass substrate (500 μm)

**Sample B**: Al (100 nm)/glass substrate (500 μm)/CP (160 μm)

**Sample C**: Al (100 nm)/glass substrate (500 μm)/NPF (2 μm)/CP (160 μm)

**Sample D**: Al (100 nm)/glass substrate (500 μm)/CP (160 μm)/NPF (2 μm)

The reflectance values at 600 nm were 84.0%, 3.4%, 3.7%, and 6.6% for Sample A, B, C, and D, respectively, as shown in Figure 8(a). Sample B had the lowest reflectance value caused by its anti-reflective CP because the light reflected from Al was linearly polarized and orthogonally rotated to 90˚ by a quarter-wave plate (QWP) in the CP and was then absorbed by the original linear polarizer axis of CP. While Sample C (with NPF on the CP) showed a relatively high reflectance value, as was expected, the reflectance of Sample D (with NPF beneath the CP) was almost the same or was very slightly higher than that of Sample B. As a result, it was found that the drop of the contrast ratio after applying NPF below the CP could not be minimal under bright outdoor conditions, but
the black color slightly faded, like the off-state mode of plasma display panel, for both Samples C and D, because Samples C and D with NPF were made with a 40% haze level, as shown in Figure 8(b). A study for improving the faded black tint effect caused by the scattering effect is in progress.

4. Conclusions

The applicability of the proposed nanoporous polymer film (NPF) for the suppression of the viewing angle dependence in the organic light-emitting diode (OLED) display application was investigated. As a result, it was found that the proposed NPF has good performance in suppressing the viewing angle dependence without a significant pixel blur phenomenon. Especially, the angular color shift of the device with NPF was reduced by about 42% compared with the pristine device. In addition, the angular luminance distribution was enlarged towards the Lambertian distribution direction. Very interestingly, the reflectance difference was minimal when NPF was embedded below the CP. Based on the study results, it was found that the proposed NPF could be a good solution for suppressing the viewing angle dependence without causing serious problems.

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