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# Directed emissive high efficient white transparent organic light emitting diodes with double layered capping layers

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ABSTRACT

We report on white transparent organic light emitting diode (TOLED) with double layered capping layer (CL), 1,1-bis[(di-4-tolylamino)phenyl]cyclohexane (TAPC) and ZnSe. By introducing the double layered CL, total efficiency was improved by 27% to have 90% of that of conventional bottom-emissive device. The achievement of highly improved efficiency was interpreted as the synergetic effect of constructive interference and maximized reflectance of CL. By adjusting the reflectance of the CL, it was possible to choose the emissions direction selectively and achieve spectral matching between top and bottom emissions as well as enhanced total efficiency in white TOLED.



## 1. Introduction

Transparent organic light-emitting diodes (TOLEDs) have been intensively investigated because they have very attractive features for next generation lighting sources which can be integrated into architectural windows, aesthetic light sources as well as see-through displays [1,2]. However, to realize practical TOLEDs a couple of technical problems have to be resolved. The total efficiency of TOLED, at best, currently reach 70% of corresponding bottom emissive OLEDs. The efficiency of TOLEDs should be improved to be comparable to that of the conventional bottom emissive OLEDs. In addition, due to the asymmetric structure of light propagation, the spectra of top and bottom emission are different. Especially in white TOLEDs, matching the spectra of bottom and top emissions is a technical challenge.

According to the several reports, the key characteristics of TOLEDs or top-emitting OLED (TEOLED) such as external quantum efficiency (EQE) and spectral distribution can be effectively modulated and improved by introducing dielectric layers or capping layers (CLs) on the outermost surface of the cathode [2–5]. As an effort to enhance the total emission of TOLEDs, majority of researches have been focused on improving the transmittance of top contact by using CLs [6–9]. However, reports on the augmenting the emission selectively to one face in white TOLEDs with CLs are scarce. Regarding white TOLEDs which are potentially important lighting sources, it is meaningful to maximize emission by collecting light into one-side of TOLED, and simultaneously to achieve high efficiency comparable to that of bottom emissive OLED. Realizing such high efficiency TOLEDs, which can selectively emit light to one face, opens the possibility of TOLED window type general lighting. During the daylight time the TOLED panel can be used as an ordinary window. After sun set the light emission direction of TOLED can be selectively directed toward to the interior of the building.

In this work, we propose white TOLED with 1,1-bis[(di-4-tolylamino)phenyl]cyclohexane (TAPC) ($n = 1.65–1.8$ in visible range) and ZnSe ($n = 2.6–2.9$ in visible range), double layered CL which can function as a reflector. We have tuned the reflectance of CL to modulate the emission

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directions and the top and bottom spectra. Both experiments and simulations have been used to investigate effects of CL thicknesses on the optical characteristics of white TOLEDs. As a result, solely due to an optical effect, enhanced total EQE and modulated emission direction and spectra were achieved.

## 2. Experiments

In order to obtain and verify effects of CLs in white TOLEDs, TOLED and conventional bottom emissive OLED structures were fabricated as shown in Fig. 1 and the highest total efficiency of those devices were compared. The TOLED (device 1) has the following stack; glass/indium tin oxide (70 nm)/TAPC (55 nm)/TCTA-Firpic (5 nm)/TCTA-Ir(2-phq)3 (1 nm)/(DCzPPy) -Firpic(5 nm)/BmPyPB (55 nm)/LiF (1 nm)/Al (1.5 nm)/Ag (15 nm)/TAPC (TAPC, 0, 20, 40, 60, 80, 100, 120, 140 nm)/ZnSe (50 nm). The thickness of the ZnSe was fixed as 50 nm. The choice of the 50 nm thickness of the ZnSe is based on our preliminary simulation results (see Supplementary). In bottom emissive OLED (device 2), we have placed a relatively thick Al metal cathode of Al (100 nm) instead of thin cathode of Al (1.5 nm)/Ag (15 nm) without CLs. Other OLED components were kept identical to those of device 1. EL emission characteristics were investigated with experimental and simulated result as a function of the TAPC thickness. Table 1 summarizes the full names of chemicals and theirs functions.

The fabrication processes have been described in our previous works [10]. Prior to the deposition ITO was treated by oxygen plasma to remove contaminates. The base pressure of all deposition processes were below $6.66 \times 10^{-5}$ Pa. All organics were deposited sequentially without breaking vacuum. The active emission area was $2 \times 2$ mm$^2$. The fabricated OLEDs were transferred directly from vacuum into an inert environment glove-box, where they were encapsulated using a UV-curable epoxy, and a glass cap with a moisture getter. The electroluminescence spectrum was measured using a Minolta CS-2000. The current–voltage-luminescence characteristics were measured with a source/measure unit (Keithley 238) and a Minolta CS-100. Transmittance of the glass cap-encapsulated TOLED was measured using an UV–visible spectrophotometer (U-3501, Hitachi). Simulations were performed using an OLED optical simulator, SimOLED [11–13]. In order to obtain realistic simulation results, we used all measured optical constants ($n$, $k$) of organic materials, which were measured using an ellipsometer (M-2000D, J.A. Woollam Co.).

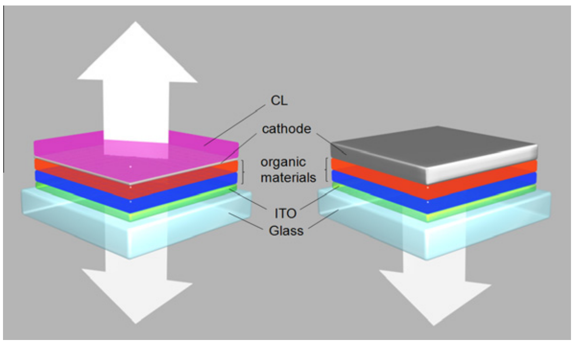


**Fig. 1.** Structures of TOLED (device 1, left) and conventional bottom-emissive OLED (device 2, right).

## 3. Results and discussion

Fig. 2a presents the variations in simulated and experimental external quantum efficiencies (EQEs) as a function of the TAPC thickness. EQEs were evaluated by integrating angular and spectral radiant intensities. Bottom, top and total values are plotted. Remarkable similarity between the simulated and measured EQEs can be observed in their EQE dependency on the CL thickness. In the device, the EQE dependencies of top and bottom emissions on the CL thickness are different. The EQE of bottom emission gradually increases as CL thickness increases up to CL of 40 nm and then decreases. On the contrary, behavior of the top side is opposite. The difference in EQE dependency on CL thickness lead to maximum emission ratio of bottom to top(B/T) of higher than 10. The highest total EQE was obtained as 12.1% at 20 nm of TAPC/ZnSe (50 nm). The EQE of TOLED without CLs was around 9.8%. This corresponds to emission improvement of 27% by using CLs. As marked as dotted line in experimental part of Fig. 2a. The EQE of conventional bottom emissive white OLEDs of device 2 is 13.4%. Thus, the total maximum efficiency of device 1 are 90% of that device 2. Fig. 2b shows the normalized radiance dependency on the viewing angle of TOLEDs with TAPC of 20 nm. The normalized radiance values of bottom and top emissions are retained up to 90% of their 0° radiance values. These features indicate that the TOLED emission characteristics of the both sides are close to Lambertian. These results demonstrates that the total efficiency of TOLEDs can be improved to the level of conventional bottom emissive OLEDs by using CLs. From the Fig. 2a, it was also found that the overall behaviors of total and the bottom EQEs show close resemblance, while the top EQE are almost opposite with much lower values. This feature indicates that the bottom emission is dominating the overall EQE.

Compared to the single TAPC case, introduction of ZnSe on TAPC brought forth an opposite effect in bottom and top emission [14]. The double CLs mainly boost the bottom emission, leading to high values of B/T and selective emission from the bottom-side. The reason of this is thought to have its origin in the reflection of the generated light toward the bottom direction.

In order to analyze optical phenomena in our device on TOLED with double layered CLs, we consider our four-layered stack of organic layer, Ag layer, TAPC layer, and ZnSe (50 nm) (Fig. 3a). Although very thin, we have considered the Ag film as an optical component. In this layout, double layered CL can be regarded as a dielectric mirror, which consists of a stack of layers with low and high refractive indices. Fig. 3a gives the schematics of various optical components and their traveling directions. In our TOLEDs, maximum bottom-side emission was achieved at a CL

**Table 1**
The full name of organics and their functions.

| Abbreviation | Chemical name | Function |
|---|---|---|
| TAPC | 1,1-Bis[(di-4-tolylamino)phenyl]cyclohexane | Hole transport layer capping layer |
| DCzPPy | 2,6-Bis(3-(carbazol-9-yl)phenyl)pyridine | Emitter host |
| BmPyPB | 1,3-Bis(3,5-di-pyrid-3-yl-phenyl)benzene | Electron transport layer |
| TCTA | 4,4′,4″-Tris(*N*-carbazolyl)-triphenylamine | Emitter host |
| Firpic | Iridium(III)bis(4,6-difluorophenyl)-pyridinato-N,C$^{2\prime}$)picolinate | Emitter dopant (blue) |
| Ir(2-phq)3 | Tris(2-phenylquinoline)iridium(III) | Emitter dopant (red) |

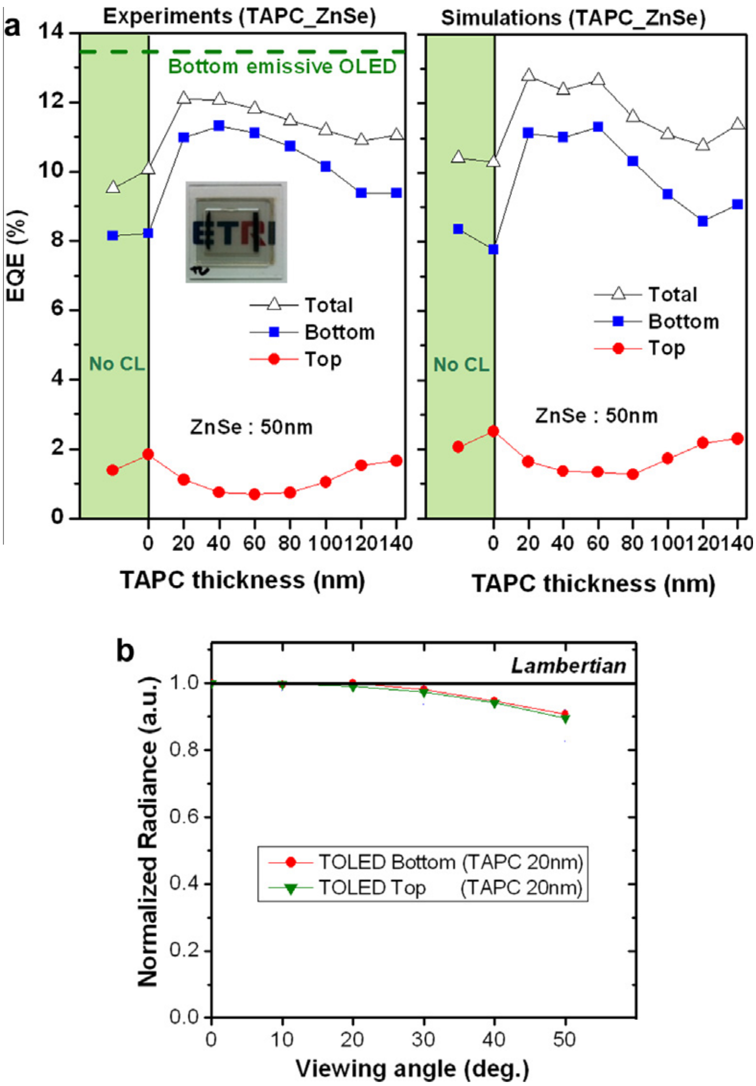


**Fig. 2.** (a) Experimental and simulated EQE in white TOLED as a function of thickness of TAPC in device 1 (The current density was kept constant as 10 mA/cm$^2$ in all EQE measurements.). (b) Normalized radiance dependency on viewing angle in TOLED with TAPC of 20 nm.

thickness of 20–60 nm. The integral bottom emission is due to the contribution from the light wave ($^{D}E_{O}$) which travels toward the bottom side directly from the light emitting layer, and $^{I}E_{R}$, $^{II}E_{R}$ and the $^{III}E_{R}$, which have been reflected from the interface I(OLED/TAPC), interface II(-TAPC/ZnSe), and interface III(ZnSe/Air), respectively.

The maximum bottom emission is achieved when the reflected components of $^{I}E_{R}$, $^{II}E_{R}$, and $^{III}E_{R}$ have mutual phase differences, which yield constructive interferences. In this case, the integral optical interaction appears as maximum reflectance of light toward the bottom face. Transmission matrix method was used to calculate trans-

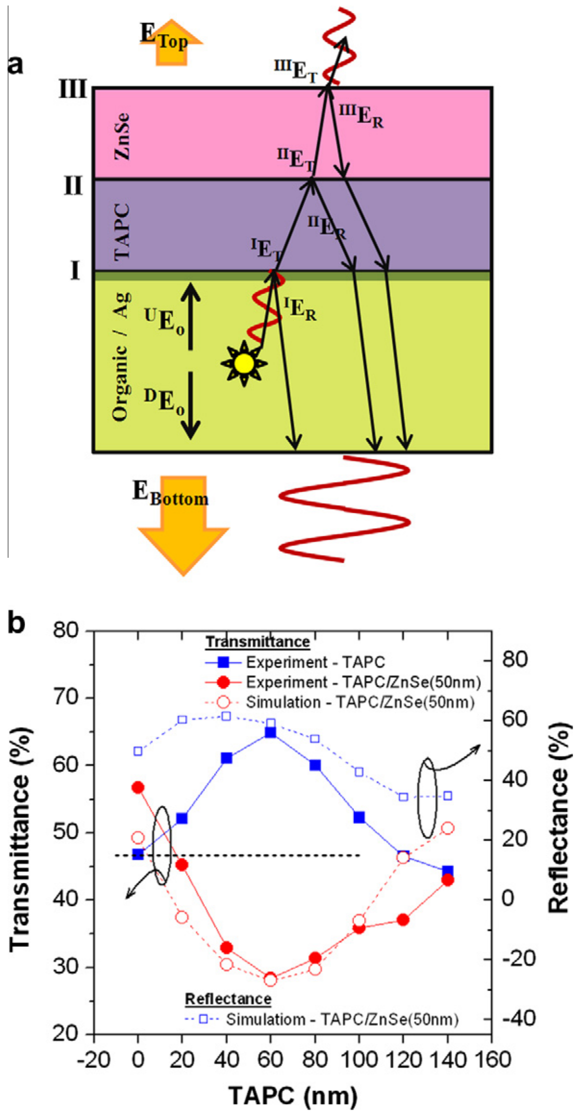


**Fig. 3.** (a) Schematics of optical components in TOLED with double layered CL. (b) Experimental and calculated transmittance averaged in the visible light range as a function of TAPC thickness.

mittance in multiple interfaces of our device [15]. This method combines Fresnel formula and Maxwell's continuity conditions to extract transmittance when the structure of consideration has multiple stacks of optical components (see Supplementary for details).

The Fig. 3b shows the calculated transmittance using the transfer matrix method, which is embedded in the SimOLED program. The overall transmittance variation trend matches the experimentally measured variation. Using the four-layered stack of organic layer, Ag layer, TAPC layer, and ZnSe, it was not possible to reproduce the tranmittance variation as a function of the TAPC thickness but also correctly calculate the lowest transmittance value. In accordance with the above results, the introduction of ZnSe (50 nm) CL on the TAPC layer increases the reflectance of the TOLED, while single CL of TAPC has an effect of increasing the transmittance. The highest reflectance of device was obtained in simulated data at a TAPC thickness of 20–60 nm where bottom-emission was maximized, as shown in Fig. 2. At these thicknesses light reflection toward the bottom side is maximized. This result strongly supports that the additional ZnSe layer on the TAPC layer is mainly acting as a reflecting component, which results in enhancement in bottom emission. Technically, we have shown the possibility of selectively choosing emission direction of TOLEDs by an optical method.

Fig. 4 summarizes the effect of introducing CLs on the electro-luminescence (EL) spectra and the color coordinates. The introduction of CLs causes the EL intensity ratio of blue to orange to vary significantly (Fig. 4a). Here two points are to be noticed. One is the change in the full width half maximum (FWHM) of blue peaks; another is the shift in orange peaks, indicating the presence of strong microcavity effects. Among them, significant shift in orange peak is noticeable, which is toward longer wavelength in bottom side and toward shorter wavelength in top side. This changes result in variation in x coordinate, toward larger x for bottom direction and smaller x for top direction as seen in Fig. 4b. As seen in Commission internationale de l'éclairage (CIE) color coordinates of Fig. 4b, spectral matching of bottom and top emissions were obtained at 40 nm TAPC. This corresponds to the CL thickness in which the maximum total emission was obtained.

Fig. 4c shows the modulated spectra of bottom and top by CL effect. Without any CLs, huge spectral deviation between bottom and top emissions is observed at greenish blue ($\lambda \sim 470$ nm). By introducing CLs, it was possible to reduce the deviation remarkably and achieve conditions which yield very low spectral difference between top and bottom emissions. Without CL the difference top and bottom coordinates was $\Delta$ (0.0747, 0.269). The relatively large disparity in y-coordinate indicates that greenish blue is the main cause of color disparity in our TOLEDs. Almost identical spectra with $\Delta$ (0.011, 0.014) were obtained when double layered CL with TAPC thickness of 40 nm were used. In our work spectral matching was achieved by adjusting the greenish blue spectrum, which resulted in reduced difference in y-coordinate.

## 4. Summary

In summary, we have fabricated TOLEDs with double layered CL of TAPC/ZnSe. With the use of TAPC/ZnSe CL it was possible to improve the total efficiency by 27%, which is the 90% of conventional bottom-emissive OLEDs. This implies that we can fabricate high efficient TOLEDs comparative to that of bottom-emissive OLEDs. By controlling the reflectance of CL, it was possible to control the emission direction selectively. Furthermore, bottom and top matching spectra were observed near the thickness of maximum total emission. We expect that spectra tunable high efficient white TOLEDs can be achieved using double CLs as an effective yet simple tool in improving and modulating the white TOLED performance.

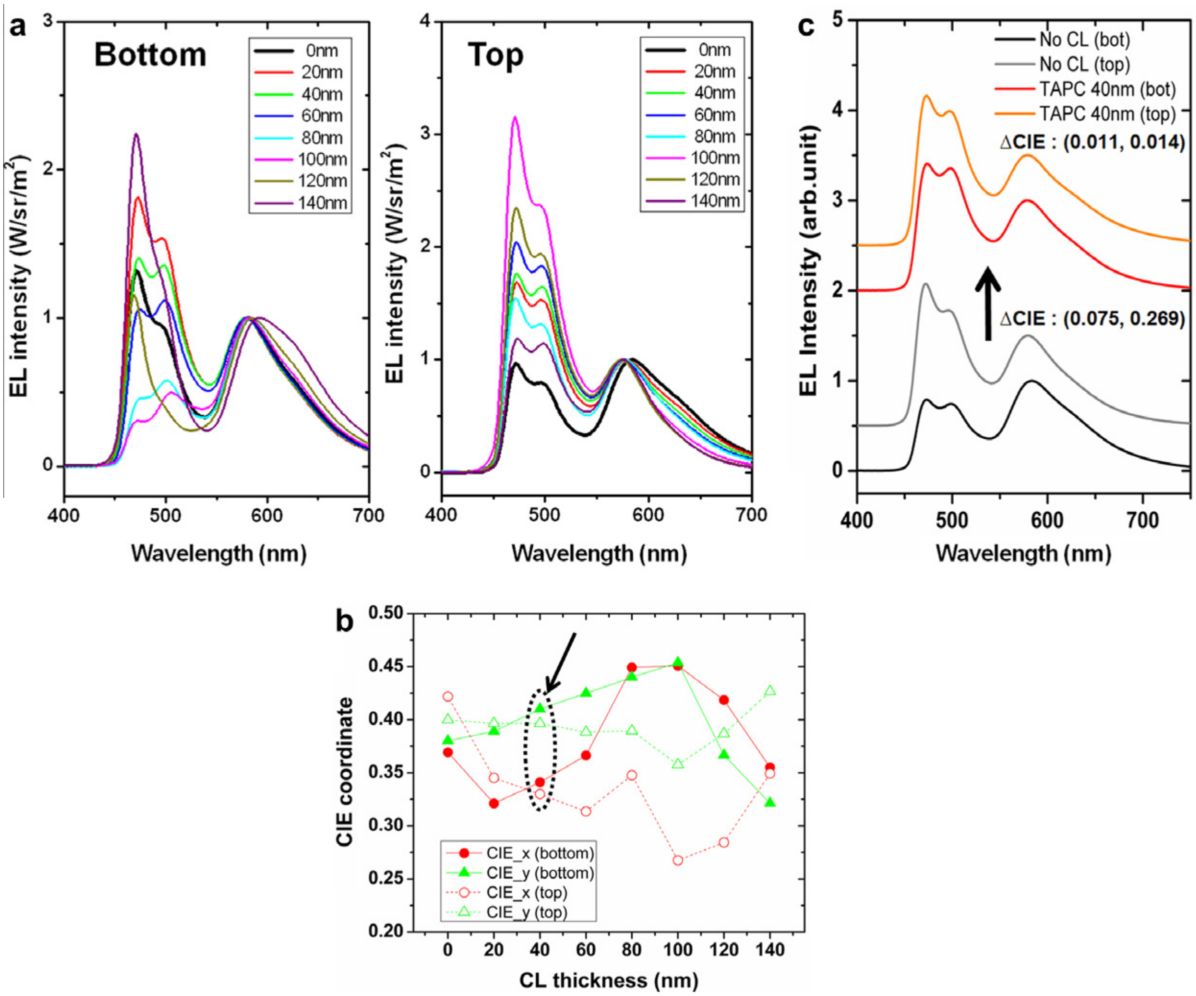


**Fig. 4.** (a) EL spectrum in bottom and top sides. (b) CIE coordinate variations in the device 1 as a function of TAPC thickness. (c) Bottom and top matched EL spectra with TAPC thickness of 40 nm in device 1 (The spectra were normalized at 580 nm peak and the spectra without CL layers were compared with these spectra. Greenish blue peak has a wavelength of $\lambda$ = 471 nm.). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

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## Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at http://dx.doi.org/10.1016/j.orgel.2012.04.005.

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