# High-efficiency microcavity top-emitting organic light-emitting diodes using silver anode

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Top-emitting organic light-emitting diodes (TOLEDs) employing highly reflective Ag as anode and semitransparent LiF/Al/Ag as cathode were fabricated. The hole injection efficiency of Ag anode can be significantly improved with surface modification using a $CF_4$ plasma. With C545T-doped $Alq_3$ emitter, the top-emitting device shows a low turn-on voltage of 2.65 V. The optimized microcavity TOLED shows a current efficiency enhancement of 65% and a total outcoupling efficiency enhancement of 35%, compared with a conventional OLED. No color variation was observed in the forward 140° forward viewing cone. Strong dependence of efficiency on Ag cathode thickness was observed, in good agreement with numerical simulations. *© 2006 American Institute of Physics*. [DOI: 10.1063/1.2172734]

Active-matrix organic light-emitting diode (AMOLED) displays have been demonstrated with high information content.[1,2] In the design of AMOLEDs, a top-emitting structure is preferred because in a top-emitting organic light-emitting diode (TOLED), the thin-film transistors can be buried under the organic light-emitting diode (OLED).[3] Thus, complicated pixel circuits can be fabricated without diminishing the aperture ratio. A TOLED can have the anode in contact with the substrate as in a regular bottom-emitting OLED, or it can have the cathode in contact with the substrate in an inverted structure. Most of the reported TOLED adopt the former structure with indium-tin oxide (ITO) as the anode on highly reflective metals, such as ITO/Ag[4] or ITO/Al.[5,6]

From a processing point of view, for such devices, it is better to have a metal as the anode in a TOLED. Recently, Wu *et al.*[7] reported a bright TOLED using a silver-silver microcavity with emission from $Alq_3$. However, the electrical and optical characteristics of such Ag anode TOLEDs were still poor as compared to conventional ITO anode devices.[8] More recently, Li *et al.*[9,10] demonstrated a very bright and efficient flexible TOLED and a white TOLED using the $CF_x$-coated Ag as an efficient reflective anode. Those results suggest that $CF_x$ coating can be more suitable for silver anode modification. Our previous research indicated that a ultrathin $CF_x$ layer can also be formed by $CF_4$ plasma pretreatment, which can significantly enhance the hole injection ability of Ag, even better than that of ITO.[11] In this letter, we report a very efficient microcavity TOLED utilizing this approach.

Apart from effective anode modification, we shall also report a microcavity where the emission spectrum is almost independent of the viewing angle. This is achieved through careful optimization and numerical modeling of the microcavity. With the use of C545T-doped $Alq_3$ instead of $Alq_3$, the full width at half maximum of the emission spectrum is about 50 nm. This narrow spectrum is also partly responsible for the weak dependence of emission wavelength on the viewing angle. Experimentally, the spectral peak shifts by only 6 nm from 0° to 70°, in good agreement with numerical simulations. It is also in agreement with the theoretical results by Djurišić *et al.*[12] that it is easier to obtain weak color shift for emitter with narrower emission spectra.

For the top cathode, LiF/Al/Ag is a good choice and is commonly used.[7,8,13] TOLEDs using such semitransparent metallic cathode should have a strong microcavity effect.[14–17] Obviously, the optical properties, such as reflectivity and transmittivity, of it is important in determining the microcavity effect. So far, few reports address the effect of the thickness of the semitransparent metal film. In this letter, we report the influence of the thickness of Ag cap layer on the overall efficiency of the TOLED. There is indeed a strong dependence and good agreement with theoretical calculation is obtained.

To fabricate the devices, a patterned 80 nm thick Ag film was first deposited on glass substrate through a shadow mask, and then pretreated under $CF_4$ plasma.[11] After surface pretreatment, the sample was loaded into an evaporation chamber at a base pressure of $7 \times 10^{-7}$ Torr for organic film deposition. The organic multilayer structure sequentially consists of alpha-napthylphenylbiphenyl diamine (56 nm) as a hole transport layer, tris-8-hydroxyquinoline aluminum [($Alq_3$) 25 nm] doped with 1 wt % 10-(2-benzothiazolyl)-1,1,7,7-tetramethyl-2,3,6,7-tetrahydro-1H,5H,11H-[1]benzo-pyrano[6,7,8-ij]quinolizin-11-one (C545T) as an emitting layer, and undoped $Alq_3$ (30 nm) as an electron transport layer. The sample was then transferred to another chamber without breaking the vacuum for cathode deposition. To achieve both desired optical transmission and effective electron injection, we applied multiple functional layers consisting of LiF(0.8 nm)/Al(2 nm)/Ag($x$ nm). The 2 nm thick Al film on LiF on $Alq_3$ is sufficient to enhance the electron injection with a negligible effect on the optical properties of the cathode.[13] The cap Ag thickness was varied from 12 nm to 50 nm.

The structure of the TOLED is shown in the inset of Fig. 1(a). For comparison, a bottom-emitting OLED with the same layer structures except that the anode is replaced by a 75 nm thick ITO and the cathode is replaced by LiF(0.8 nm)/Al(120 nm) was also fabricated. Another control device using as-deposited Ag without $CF_4$ pretreatment was made as well. Current density-voltage ($J$-$V$) characteris-

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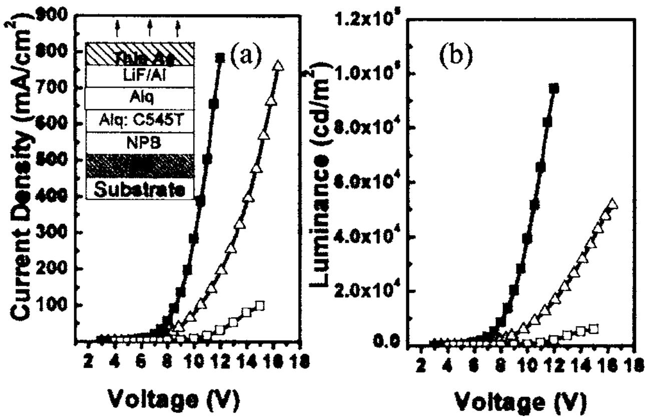


FIG. 1. (a) $J$-$V$ and (b) $L$-$V$ of various devices. Conventional bottom-emitting OLEDs (△); TOLEDs using pretreated Ag (■); and as deposited Ag (□). The top Ag cathode has a thickness of 20 nm for both TOLEDs. Shown in the inset is the TOLED configuration.

tics of the devices were measured using an Advantest R6145 dc source and a Fluke 45 dual-display multimeter. The spectral characterization and electroluminescence (EL) intensity were measured using a PR650 SpectraScan spectrophotometer. The measurement and data acquisition were controlled by National Instrument's LabWindows software.

Figure 1 compares the current density ($J$)-normal direction luminance ($L$)-voltage ($V$) characteristics of the references OLED and the TOLEDs. The voltage drop on the electrode lines were subtracted for better comparison. The TOLED using as-deposited Ag anode shows rather poor performance. On the other hand, the TOLED using surface-modified Ag anode demonstrates performances superior to that of the control bottom-emitting device. For instance, the TOLED has a turn-on voltage as low as 2.65 V, compared with 2.85 V for the OLED to obtain a brightness of 1 cd/m$^2$. The voltages to obtain a luminance of 1000 cd/m$^2$ for the two devices are 6.1 and 7.0 V, respectively.

Both the $J$ and $L$ of the pretreated Ag anode TOLED increase faster than those of the other OLEDs with increasing voltage. The superior $J$-$V$ characteristic is attributed to the enhanced hole injection of the Ag anode with $CF_4$ plasma pretreatment. It is proposed that the hole injection barrier is effectively reduced by the formation of a localized interface dipole layer with the deposition of an ultrathin $CF_x$ layer on the Ag anode surface.[11] The difference between the $L$-$V$ curves of TOLED and bottom-emitting OLED is bigger than that between the $J$-$V$ curves. This is due to a strong microcavity effect. Figure 2 shows the current efficiency $\eta_J$ as a function of current density of the bottom-emitting OLED and the TOLED using 20 nm Ag cathode. At 10 mA/cm$^2$, the TOLED demonstrates a high efficiency of 16.1 cd/A, 65% higher than that of the control OLED. For the rest of this letter, we shall not deal with non-treated Ag anode. TOLED refers to the devices using treated Ag as anode.

It is expected that the thickness of the Ag cap on the LiF/Al cathode should affect the microcavity and hence the performance of the device. It was found that all TOLEDs with different Ag cathode thicknesses had identical $J$-$V$ curves within measurement uncertainty (not shown). However, the $L$-$V$ curves are quite different. Figure 3 shows the TOLED current efficiency in the normal direction as a function of the top Ag thickness at 10 mA/cm$^2$. It can be seen that there is a strong dependence on the Ag thickness. The

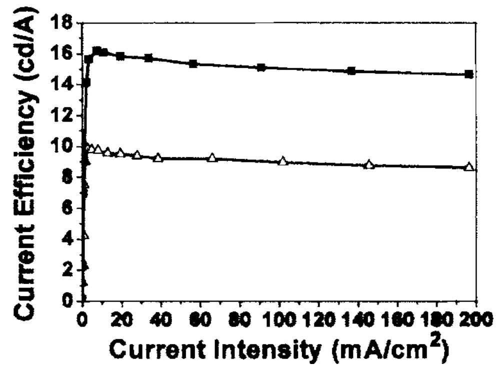


FIG. 2. $\eta_J$-$J$ of the bottom-emitting OLEDs and TOLEDs using pretreated Ag as anode.

absorption of the Ag in this range actually differs by less than 2%, according to the calculation results shown in inset of the same figure. Therefore, the difference in efficiency is attributed to the changing reflectivity of the Ag films.

The emission characteristics of TOLEDs can be accurately described by a model based on dipole radiation in a multilayer thin-film structure.[18,19] In the normal direction, the emission intensity is given by

$$I(\lambda) \propto \frac{|1+\sqrt{R_1}\exp[j(4\pi n_0 z/\lambda+\phi_1)]|^2}{|1-\sqrt{R_1R_2}\exp[j(4\pi n_0 d/\lambda+\phi_1+\phi_2)]|^2}T_2(\lambda), \tag{1}$$

where $R_2$ and $T_2$ are the respective reflectance and transmittance of the top semitransparent cathode; $R_1$ is the reflectance of the bottom opaque anode; $\phi_1$ and $\phi_2$ are the respective phase changes at the two interfaces, and $n_0$ is the refractive index of the medium between the electrodes; $z$ is the distance between dipole and Ag anode; $d$ is the cavity length; and $\lambda$ is the emission wavelength in a vacuum. Based on Eq. (1), with increasing Ag cathode thickness, the transmittance $T_2$ decreases while the reflectance $R_2$ increases, which leads mathematically to a maximum value of the output intensity at a Ag cathode thickness of about 24 nm. The solid line in Fig. 3 shows the calculation result. It can be seen that the agreement with experimental data is excellent. These results indicate that the thickness of the semitransparent top layer plays an important role in determining the device efficiency.

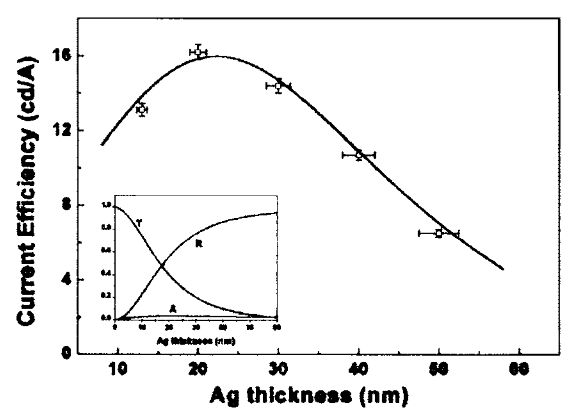


FIG. 3. Current efficiency in the normal direction vs thickness of the Ag cathode in TOLED. The simulation results (solid line) were scaled to give the best fit to the data (square). Inset: Transmittance ($T$), reflectance ($R$), and absorption of Ag film with different thicknesses.

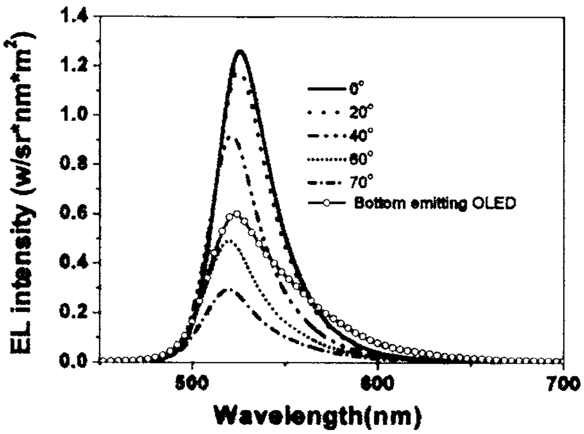


FIG. 4. Spectral characteristics of TOLED with 20 nm Ag top cathode at different viewing angles and of conventional OLED in the normal direction. All data are taken at 10 mA/cm$^2$.

Figure 4 shows the emission spectra measured under a current density of 10 mA/cm$^2$ at viewing angles from 0° to 70° off the normal direction for the TOLED using 20 nm Ag cathode. The EL spectrum of the control device is also shown for comparison. It can be seen that the Ag TOLED shows a narrower EL spectra due to microcavity effects.[15,20,21] A large spectrum shift with viewing angles was observed in previously reported microacvity TOLEDs as well as bottom-emitting OLEDs.[20,21] However, based on a narrower emission spectra emitter together with optimization of the organic layer thicknesses, the color variation is almost eliminated in the present TOLED where the spectral peak shifts by only 6 nm from the normal direction to 70°.

Figure 5 shows the relative angular distribution of EL intensity for the two devices. The bottom-emitting control device shows a Lambertian distribution, while the TOLED shows a sub-Lambertian distribution with enhanced intensity in the forward direction and suppressed intensity at the large viewing angles ($>50°$). The external quantum efficiency ($\eta_E$) of both devices are evaluated by integrating the photon flux using the formula $\eta_E = 2\pi/J\Sigma I(\theta)\sin(\theta)\Delta\theta$, where $I(\theta)$ is the measured photon radiation intensity at angle $\theta$. The spatial integration results indicate that $\eta_E$ of Ag-anode device is about 35% higher than that of the bottom-emitting device. This result indicates that the TOLED is more efficient than the corresponding bottom-emitting OLED, due to improved light coupling efficiency. The electro-optical characteristics of bottom-emitting OLED and optimized TOLED are compared in Table I. It is clear that the Ag TOLED demonstrates better performance as compared to the control bottom-emitting device.

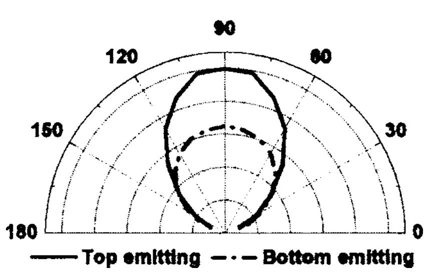


FIG. 5. Angular distribution of radiation intensity for the TOLED and a conventional OLED.

TABLE I. Comparison of the characteristics of a new TOLED with a conventional OLED.

| Device | $V_{\mathrm{on}}$ @ 1 cd/m$^2$ (V) | $\eta_{J\max}$ (cd/A) | $\eta_{E\max}$ (%) | $\eta_J$ @ 1000 cd/m$^2$ (cd/A) | $\eta_p$ @ 1000 cd/m$^2$ (lm/W) |
|---|---|---|---|---|---|
| TOLED | 2.65 | 16.3 | 3.6 | 16.1 | 5.52 |
| OLED | 2.80 | 9.7 | 2.7 | 9.6 | 4.12 |

In conclusion, efficient top-emitting OLEDs have been fabricated using highly reflective Ag as the anode. Surface modification of the Ag anode by $CF_4$ plasma substantially enhances the hole injection efficiency. The color variation is almost eliminated in the present TOLED. Besides, the thickness of the top Ag layer was found to play a key role in determining the device efficiency. The optimized microcavity TOLED has a current efficiency enhancement of 65% and a total outcoupling efficiency enhancement of 35%, as compared with a conventional OLED.

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