Ultrathin Cu-Ag Anode for High Light Outcoupling Efficiency by Eliminating Waveguide Mode in OLED

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Abstract
Extracting light from the waveguide mode in organic light-emitting diodes (OLED) is most challenging as such optical mode is intrinsically formed due to materials and layered structure used in the device. Here, we show that a metal-only transparent conductor based an ultrathin Cu-Ag film can be used to eliminate the guided mode, thus enhancing light extraction efficiency in OLED.

Author Keywords
Organic light-emitting diodes (OLEDs); metal-based transparent conductor; anti-reflective; optical outcoupling; waveguide mode.

1. Objective and Background
Organic light emitting diodes (OLEDs) are being widely used as emitting cells in flexible and transparent display products (1). Despite the success in material progress and achieving 100% internal quantum efficiency (IQE), external quantum efficiency (EQE) of OLED is still limited due to the poor outcoupling efficiency (2). Therefore, searching for solutions to increase the EQE of OLEDs has been the focus of this research area.

There are three major factors that limit the EQE of OLEDs: (1) generated photons in a device are lost to the contact metal as plasmonic mode and (2) internally reflected back from a glass substrate, and (3) light is trapped in a device in the form of guided mode due to the device layers serving as waveguide (2, 3). There have been many reports on means to suppress plasmonic mode or efficiently extract trapped light from the substrate (4-6). However, extracting or minimizing the guided mode formed within the active region including the transparent anode still remains a challenge as such optical mode is created inherently and propagates along these layers. Especially when sufficiently thick layer of transparent conductive oxides (TCO) as an anode is needed to ensure low film resistivity, waveguide mode in this high refractive index material becomes inevitable.

There have been efforts to extract the waveguide mode. But all of them use patterning or grid, which is easy to protrude into organic layers or results in non-planar surfaces, negatively affecting the surface smoothness (6, 7). Also, these methods are never a favorable solution since several additional fabrication steps are required that increases the fabrication cost. In this work, we introduce a simple yet effective solution to eliminate the waveguide mode and increase EQE without adding fabrication cost or affecting other properties of the OLEDs.

Our approach is to use an extremely thin Ag layer, less than 6 nm, to replace the traditional ITO electrode. This makes the entire OLED stack thinner than 160 nm, which helps to suppress waveguide mode in the OLED structure and thereby enhance its EQE. We show that a complete elimination of waveguide mode is possible only through using our extremely thin metal which is never possible by conventional TCO counterparts such as ITO or even the widely exploited dielectric-metal-dielectric (DMD) based TC that involves dielectric layers with high refractive index material (8, 9). This finding opens up a new opportunity to harness the benefit of metal-only transparent conductor (TC) as a means to achieve high performance OLEDs through efficient light outcoupling, which has not been possible with other types of TCOs. Moreover, our solution is not only simple in process but also can achieve high throughput with excellent compatibility with existing OLED manufacturing process.

2. Results and Discussion
Seed layer effect on Ag film growth: The key to metal-based TC lies in controlling metal film's thickness well below the skin depth so that high light transmission can be achieved. Ag is known to have superior electrical conductivity and low optical loss at visible wavelength. However, Ag films tend to grow in 3D island-like (Volmer-Weber) modes on dielectric substrates leading to high electrical resistance and optical scattering loss (10). With appropriate seed-layer, we found that the Ag layer can be controlled to have 2D-like growth forming a smooth and continuous film with low electrical resistance and optical loss (11). First, optimum thickness of Cu as a seed layer was investigated by varying Cu thickness from 1-20 Å, followed by the subsequent deposition of 4-5 nm thick Ag. By measuring sheet resistance (R_S), average transmittance (T_{AVE}), and surface roughness, the effect of seed layer thickness on Ag film as a TC was studied as shown in Fig. 1. Since Cu is optically lossy at visible wavelength, too thick of a seed layer can compromise the T_{AVE} while too thin of a layer is insufficient to form high density of nucleation sites for Ag to form a continuous film.

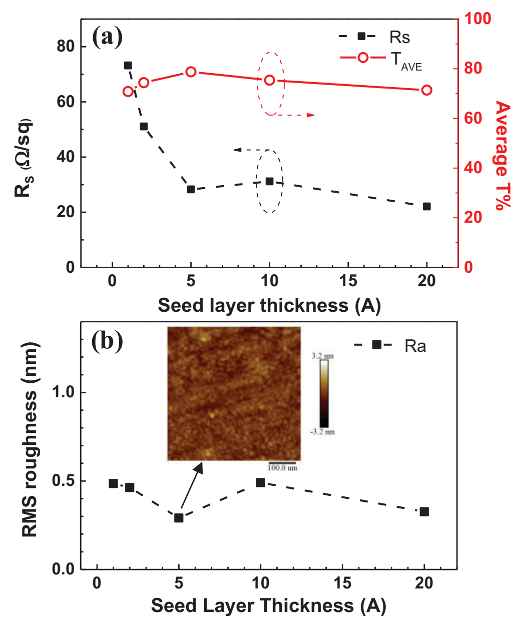

Figure 1. (a) R_S, T_{AVE}, and (b) root-mean-square (RMS) surface roughness for 4-5 nm of Ag film with varying seed layer thickness. Inset: AFM of Ag film with 5 Å of Cu-seed.

Based on the performance of Ag film, the optimum thickness of Cu seed-layer was 5 Å, which guarantees ultrathin and smooth Ag film with low electrical resistance and high transmittance.

Thickness dependent property: Based on the optimum thickness of Cu-seed layer, Cu-Ag film's electrical and optical properties are studied in comparison with bare Ag film to show the benefit of Cu-Ag as an excellent candidate for extremely thin metal-only anode material. Figure 2 (a) shows R_S as a function of film thickness (d_{Film}) for Cu-Ag film where significant reduction of Ag film's percolation threshold is observed compared with Ag film without any nucleation layer.

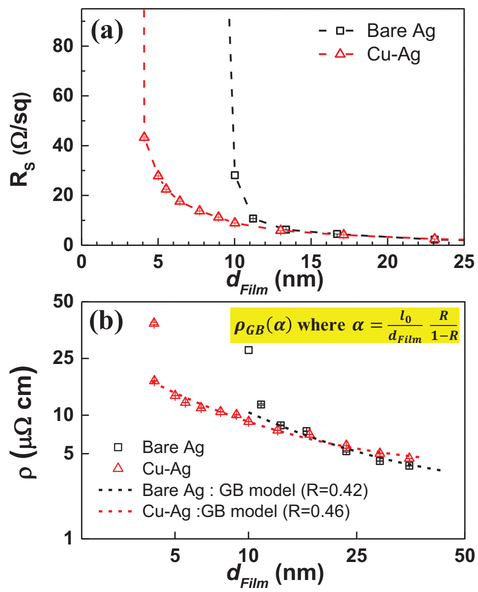

Figure 2. (a) R_S and (b) ρ as a function of d_{Film} for Cu-Ag and bare Ag film.

It is remarkable that the film can be electrically conducting even at a total film thickness as low as 4 nm. Next, Mayadas-Shatzkes (M-S) model was adopted to approximately quantify the degree of grain boundary scattering in each film as this would be an important indicator of the quality of the film(12). With excluding the surface contribution, the fitted values show that Cu-Ag film does not show significantly deteriorated reflection coefficient (R) meaning the electrical resistivity (ρ) property is not significantly compromised for Cu-Ag film compared to bare Ag even at a sufficiently thin regime.

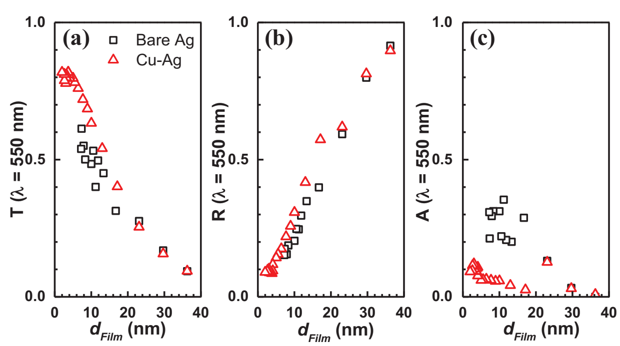

Figure 3. (a) T, (b) R, and (c) A versus d_{Film} at 550 nm of wavelength for Cu-Ag and bare Ag film.

Comparing the optical properties of Cu-Ag and bare Ag films, Cu-Ag films show higher transmittance (T) and significantly lower absorbance (A) at 550 nm of wavelength (λ) under film thickness of sub-10nm range, which is attributed to the smooth and continuous film giving minimum light absorption or scattering. At a film thickness of 5-6 nm under optimum Ag deposition condition, Cu-Ag film can have T close to 80% with A around 5-6 % with R_S as low as 20Ω/sq.

FOM and AR effect: From the electrical and optical properties of Ag based films discussed above, their performance as a TC can be calculated using Haake's figure of merit (FOM) value ϕ_{TC}, which is the ratio of optical transmittance over sheet resistance (see equation in the inset of Fig. 4 (a)) (13). The plot of ϕ_{TC} for Cu-Ag and bare Ag film as a function of d_{Film} shown in Figure 4 (a) indicates max ϕ_{TC} for Cu-Ag is at least one order of magnitude higher than that of bare Ag. Based on this FOM plot, Cu-Ag for TC application has optimum thickness at d_{Film} = 5.5 nm with T = 77% at 550 nm and R_S = 22Ω/sq. Although T is quite high for typical ultrathin metal film, the reflectance (R) is still around 15% which is quite reflective.

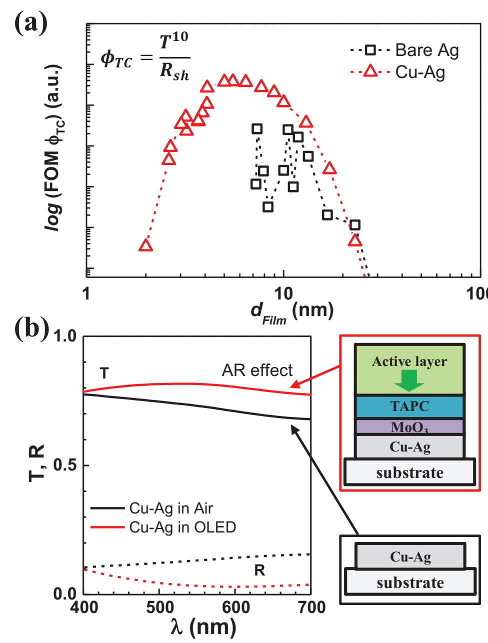

Figure 4. (a) $log(\phi_{TC})$ versus d_{Film} plot and (b) anti-reflective effect of Cu-Ag inside OLED structure.

Importantly, the reflectance of photons from the emissive layer can be significantly minimized by the use of hole transporting layer (HTL) in OLED directly adjacent to the anode, which functions as an effective anti-reflective (AR) layer. Figure 4 (b) shows the simulation result of T and R of ultrathin Cu-Ag film alone on a substrate (black) and inside the OLED structure as an anode (red) where the light path is from the emission layer (EML) through TAPC as HTL, MoO_x as hole injection layer (HIL), TC, and to the substrate. The thickness of EML, HTL, and MoO_x layers were optimized to obtain the best electrical characteristics as shown in Figure 5 (a). From Fig. 4(b), it can be deduced that the HTL and injection layer together can act as an effective AR coating for TC which enhances the transmittance of light generated from the EML to well above 80% by effectively suppressing R below 5% at 550 nm of λ. Note that such low R is close to the reflectivity of the substrate itself. Another great benefit of this strategy is that the total device thickness including the TC anode is kept at a minimum thickness level, which can suppress the confinement of waveguide mode inside the active region, i.e. the thickness can be reduced to below the cut-off thickness.

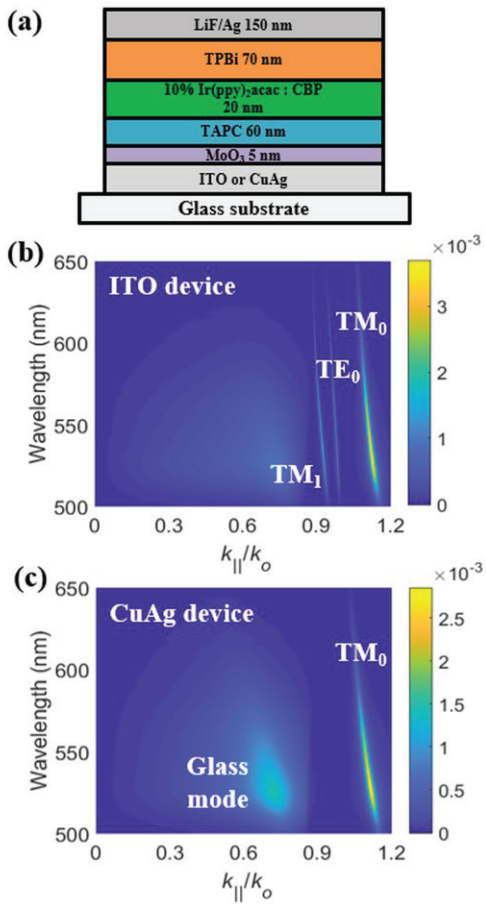

Figure 5. (a) Structure of OLEDs investigated in this work. Dispersion relations of (b) ITO and (c) Cu-Ag devices. Modal power distribution with each mode is indicated.

Dispersion relationships: ITO is used as an anode for a reference sample, and the other is replaced with Cu-Ag to evaluate the efficiency improvement. Figure 5(a) shows schematics of OLEDs fabricated in this work. The anode side is the same as discussed above and on the cathode side, LiF and TPBi are used for electron injection and transporting layers, respectively. Emissive layer is 10% $Ir(ppy)_2acac$ doped in CBP matrix and located at the center of the devices. Glass substrate is used for both ITO and Cu-Ag devices for better comparison.

Figures 5(b) and (c) show dispersion relations of ITO and Cu-Ag devices, respectively. Dipole simulation was conducted with OLEDs to investigate existing modes in the structure (14). Dipole was placed at the center of the EML and permittivity of materials used for calculation was all measured by ellipsometry.

Figure 5(b) is the dispersion relation of ITO device. It shows large portion of plasmonic mode (TM_0) associated with the cathode metal, and two waveguide modes (TE_0 and TM_1) associated with the active layer and ITO. This implies that electrical luminescence at the EML intrinsically couples to two waveguide modes that cannot be extracted by external means. Interestingly, the dispersion relation of the Cu-Ag device does not show any waveguide modes as shown in Figure 5(c). It indicates that light excited at the EML does not create propagating mode along the organic stacks; this effect, in turn, increases the portion of light as a glass mode that can be extracted.

Mode profiles: We also obtained field profile of each mode found in the dispersion curves. We solved Maxwell's equations with respect to the 1D boundary conditions and found each mode shown in the dispersion relation plotted in Figure 5. Fields created in the ITO and CuAg devices are plotted in black and red curves, respectively in Figure 6.

Figure 6(a) shows TM_0 modes of both the ITO and Cu-Ag devices. In the figure, each field profile includes either ITO or Cu-Ag. Figures 6(b) and (c) show TE_0 and TM_1 modes created only in the ITO device. Figure 6(d) is the glass mode of the Cu-Ag device, that is oscillating in the glass substrate.

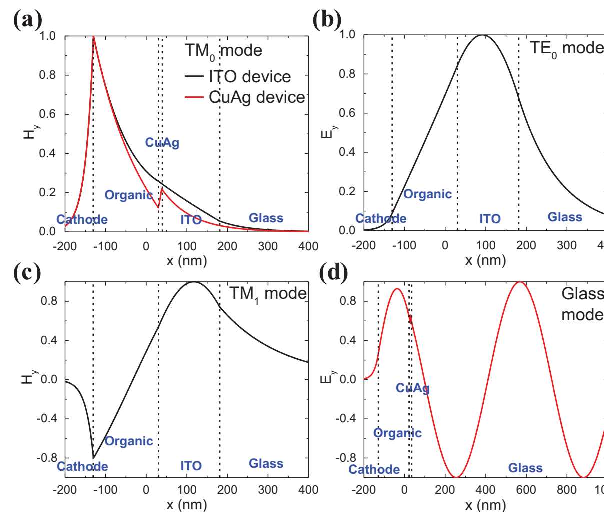

Figure 6. Field profile of each mode formed in the ITO and Cu-Ag devices. (a) TM_0 mode, also called plasmonic mode. (b) TE_0 and (c) TM_1 modes, which are waveguide modes. (d) Light trapped in glass substrate, which is called glass mode. Cu-Ag or ITO is used for calculation in (a).

As shown in Figure 6(a), TM_0 mode is the plasmonic mode that is confined significantly at the interface between organic and Ag cathode. The two waveguide modes show large field portion at the ITO anode. Glass mode exists in the ITO device too, but its portion is significantly lower than the other three modes.

The disappearance of waveguide mode in the CuAg devices increases energy portion of the substrate mode rather than plasmonic mode. As one can compare Figure 5 (c) and (d), the maximum energy portion of the plasmonic mode decreases in the CuAg device, which likely comes from the reduced confinement in the organic stacks.

Device performances: We have fabricated OLEDs shown in Figure 5(a). Since the results shown in Figures 5(b) and (c) indicate that disappearance of waveguide mode is likely to increase energy confined in the substrate mode, index-matching fluid (IMF) is used to extract the substrate mode to observe the effect of eliminating the waveguide mode.

Without any special outcoupling schemes, EQE of the reference device is calculated to be around 20%. Slight mismatch between the theoretical value and the measurement mainly stems from nonideal case of electron-hole recombination. The calculated EQE of 20% results from the assumption of 100% IQE, but it is more likely that IQE does not reach 100% in real devices. For example, the device can experience exciton annihilation at the EML and ETL interface since CBP and $Ir(ppy)_2acac$ both transport holes more favorably than electrons.

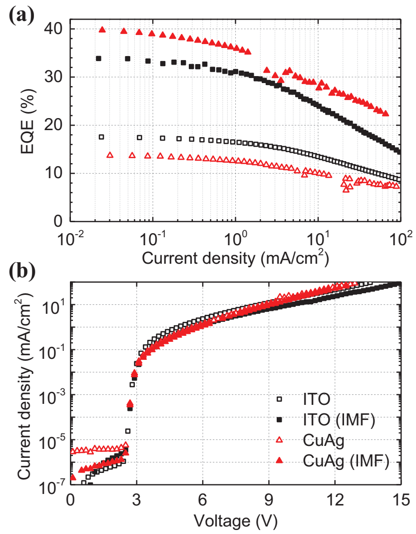

Figure 7. Device performances of ITO and Cu-Ag devices. (a) EQE and (b) current density -voltage characteristics of ITO and CuAg devices with and without IMF. IMF is used to extract light trapped in the substrate.

Since the generated photons gets absorbed in the Cu-Ag layer, EQE of the Cu-Ag device is lower than that of the ITO device without IMF. However, the EQE of the CuAg device becomes significantly enhanced when substrate mode is extracted with the help of IMF since it has more portion of light trapped in the substrate due to the elimination of waveguide mode in the active and anode layers. Overlap of the current density -voltage curve in Figure 7 (b) for ITO and Cu-Ag devices indicates that the effect of electrical injection is minimum in which we can safely conclude that the enhancement in the EQE after applying IMF for Cu-Ag is attributed to the efficient optical outcoupling.

3. Conclusion

Coupling of the emitted light to the waveguide mode represents a significant waste of photons generated from the emission layers. Unfortunately, conventional TCO is prone to this waveguide mode as its indispensably thick layer will only serve to aid this waveguide mode. In this work, we introduced a design strategy to utilize high performance Ag based metal-only TC with Cu as a seed-layer. With deliberate engineering of Cu-Ag film, we showed that extremely thin layer of Cu-Ag TC as an anode can completely eliminate waveguide mode and increase EQE without compromising any other properties of OLEDs. An ultra-thin Cu-coated Ag film less than 6 nm developed by us is highly conductive and transparent along the visible wavelength range. Moreover, by taking the advantage of AR effect from HTL, Cu-Ag's optical performance can be boosted. By replacing conventional ITO with extremely thin Cu-Ag film, the total thickness of OLEDs is maintained thinner than a cut-off thickness for the formation of an optical mode along the device, thereby suppressing the waveguide mode in the device. This simple and yet novel approach taken in this work will pave the way to replace ITO and increase efficiency in flexible and transparent OLEDs that is widely used in display industry.

4. Impact

Extraction of waveguide mode is the biggest challenge in the OLEDs community, and most of the efforts to outcouple waveguide mode has been focused on inserting patterns or grids on the ITO anode. This work introduces a novel way to completely remove waveguide mode by using extremely thin Ag film as anode. Conventional TCOs such as ITO or IZO, etc, cannot be as thin as Cu-Ag film used in this work since the thin metal oxides are substantially more resistive. Increasing the thickness of TCO can reduce the resistivity of the film but inevitably functions as a waveguide that traps the emitted light. The novelty of this work lies in the design of extremely thin Cu-Ag as an anode and the demonstration of it as an TC anode in OLED structure that could avoid formation of waveguide mode and increase light outcoupling efficiency.

5. Acknowledgements

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6. References

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