4.2: Highly Efficient Top-Emitting White Organic Electroluminescent Devices

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Abstract
We have developed highly efficient white top-emitting organic light-emitting devices (TOLEDs) with broad emission by modifying both anode and cathode. To alleviate the undesirable microcavity effect and obtain “broad” white emission, CFx-coated Ag anode with modified reflectivity and an index-matching layer (SnO₂) capped on thin Ca/Ag cathode with a maximal transparency of 80% were employed. A top-emitting broad white light device based on the dual layer architecture of light blue and yellow emitters with one of the highest EL efficiencies of 22.2 cd/A (9.6 lm/W) at 20 mA/cm² and 7.3 V with Commission Internationale d’Eclairage (CIE) coordinates of (x = 0.31, y = 0.47) was demonstrated.

1. Introduction
Organic light-emitting devices (OLEDs) [1] have been well recognized in recent years as one of the best flat panel display technologies that are capable of meeting the most stringent demand of future display applications. To realize the full potential of this display technology, full color top-emitting OLED structure (TOLED) coupled with a LTPS-TFT active matrix (AM) backplane appears to be the most attractive. This is because TOLED can provide not only higher aperture ratio (AR) than the usual bottom emitting one, but also higher display image quality that often necessitates a more complicated drive circuit in AMOLEDs. In addition, it is also well established that pixels with high AR in the panel invariably lead to prolonged operational stability owing to less current density needed to drive each pixel in order to achieve a desired luminescence. In recent years, there also has been considerable interest in developing high-efficiency white OLEDs. This is because it provides the full color active matrix OLEDs with the alternative approach using white OLEDs coupled with color filter (CF) that can circumvent the problematic shadow mask for RGB pixelation in production and achieve much higher display resolution. This led Sanyo/Kodak to declare in 2003 that white light OLEDs with on chip CF and LTPS-TFT combined with top emission architecture will be the most attractive full color OLED technology in the future [2]. However, it remains a challenge to generate white light over a broad range of the visible spectrum in top-emitting OLEDs because it is unavoidably interfered by strong microcavity effect. To the best of our knowledge, no report has been documented on generating highly efficient broadband white light emissions in a top-emitting device.

In SID 2004, Sony demonstrated a full color display with vivid images employing a “Super Top Emission” scheme which was combined with white emitter, microcavity structure and color filter array [3], in which three-wavelength white light was adjusted under specific optical length to optimize microcavity effects in the respective RGB pixels. Although a white emitter was used, individual RGB light instead of broad white emission were created by patterning three different thicknesses of ITO. In mass production consideration, the requirements of a three-wavelength white light coupled with three different and matching thickness of ITO patterned on one substrate could be difficult and not compatible with existing manufacturing process. We report herewith a new device structure by modifying electrodes, and controlling optical length of the light-emitting devices from which a top-emitting device with high EL efficiency of 22.2 cd/A and a near white light emission of Commission Internationale d’Eclairage (CIE) coordinates of (x = 0.31, y = 0.47) was achieved.

2. Microcavity
A microcavity structure in a top-emitting device is formed between a reflective anode and a semi-transparent cathode and the microcavity effect can be realized as one kind of Fabry-Perot filters which should satisfy the following equation:

\[ \frac{2L}{\lambda_{\text{max}}} + \frac{\Phi}{2\pi} = m \quad (m = \text{integer}) \quad (\text{Eq. 1}) \]

where \( L \) is the optical length between the two mirrors, and \( \Phi \) is the sum of the phase shift from anode and cathode [4].

The full width at half maximum (FWHM) [5, 6] can be estimated by

\[ \text{FWHM} = \frac{\lambda^2}{2L} \times \frac{1}{\pi \sqrt{\frac{R_1 R_2}{\sigma} R_1^2 R_2^2}} \quad (\text{Eq. 2}) \]

where \( R_1 \) and \( R_2 \) are the reflectance of the two mirrors. Spectral narrowing is the most common phenomena caused by strong microcavity effect. As implied from equation 2, shorter \( L \) as well as lower \( R_1 \) or \( R_2 \) are preferred to alleviate the undesirable microcavity effect and to obtain wider FWHM emission.

3. Device fabrication
3.1 Device architecture
In order to study the optical interference in the white light top-emitting device, three device structures are designed and fabricated as shown in Figure 1. Chemical structures of key materials used in this study as white emission are also shown in Figure 1. In device A, the structure was [Ag (200 nm)/ITO (75 nm)/ 4,4’-bis[N-(1-naphthyl)-N-phenyl-amino]biphenyl (NPB)
(50 nm)/1.5 wt% Rubrene : NPB (20 nm)/3 wt% p-bis(p-N, N-diphenyl-aminostyryl)benzene (DSA-Ph) : 2-methyl-9,10-di(2-naphthyl)anthracene (MADN) (40 nm)/ Aluminum tris(8-hydroxyquinoline) (Alq3) (10 nm)/Ca (5 nm)/Ag (15 nm), which is the conventional white light top-emitting device. In device B, ITO is the transparent anode, and cathode is Ca (5 nm)/Ag (15 nm) capped with an index-matching layer of SnO2 (22.5 nm). Organic white emitter in device B is the same as in device A. In device C, we used 200-nm-thick Ag as reflective anode coated with a polymerized fluorocarbon film (CFx) as the hole injection layer [7]. Cathode and organic white emitter were the same as in device B. Organic materials were deposited by thermal evaporation in an ULVAC Solciet OLED coater at a base vacuum of 10^{-7} Torr. All devices were hermetically sealed prior to testing. Electroluminescent (EL) spectra, luminance yield and CIE x,y color coordinates were measured by a Photo Research PR-650 spectrophotometer driven by a programmable dc source.

3.2 Anode
In top-emitting OLEDs, high reflectivity of the anode is important for achieving high luminance efficiency [8]. In our study, 200 nm Ag with a good reflectivity of more than 95% was used as anode. Furthermore, by direct photoionization measurements (Riken AC-2), the work function of Ag is found to be at 4.65 eV which is comparable to that of ITO and like which, CFx is overcoated onto the Ag surface to further improve its hole injection.

3.3 Cathode and index matching layer
Figure 2 shows transmission spectrum of Ca/Ag cathode capped with different thicknesses of SnO2 varying from 7.5, 15, 22.5 and 30 nm. We realize that transmission of metal cathode is a critical issue to be contended in TOLEDs because of its nature of low transparency. In order to improve transparency of the cathode, high refractive index (n=2.0) of SnO2 was deposited via thermal evaporation. With increasing thickness of SnO2, wavelength with optimal transmittance is shifted gradually from blue to red region. Here, 22.5-nm-thick SnO2 was selected due to its higher transparency throughout the entire visible region of our broad white emitter.

4. Result and discussion
The detailed EL performances of these devices are summarized in Table 1 and their EL spectra are plotted in Figure 3. By observing the variation in the EL spectra, we can analyze and understand more about microcavity effect caused by the device structure. It is apparent that most of white light generated in device A was whittled down and showed a narrow spectrum exhibits peak wavelength at 468 nm with FWHM of 20 nm. Saturated color with CIEx,y coordinates (0.12, 0.17) and low efficiency of 0.6 cd/A were observed. Although metal/ITO was often chosen as anode in
4.2 / S.-F. Hsu

Conventional top-emitting devices [9], the contribution of optical length is larger in ITO layer (n ~ 2.2) than that of organic layer (n ~ 1.6-1.7) and longer optical length tends to lead stronger microcavity effect in device A. To alleviate this unwanted microcavity effect, ITO layer should be eradicated, and optical length of device can be efficiently shortened without altering the recombination region within the white emitter.

Table 1. EL performance of device A, B and C measured at 20 mA/cm².

<table>
<thead>
<tr>
<th>Devices</th>
<th>Voltage (V)</th>
<th>Lum. Yield (cd/A)</th>
<th>Efficiency (lm/W)</th>
<th>CIE (x,y)</th>
<th>FWHM (nm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>5.3</td>
<td>0.6</td>
<td>0.4</td>
<td>0.12, 0.17</td>
<td>20</td>
</tr>
<tr>
<td>B</td>
<td>6.6</td>
<td>2.7</td>
<td>1.3</td>
<td>0.21, 0.35</td>
<td>64</td>
</tr>
<tr>
<td>B (bottom)</td>
<td>6.6</td>
<td>6.6</td>
<td>3.1</td>
<td>0.27, 0.40</td>
<td>116</td>
</tr>
<tr>
<td>C</td>
<td>7.3</td>
<td>22.2</td>
<td>9.6</td>
<td>0.31, 0.47</td>
<td>136</td>
</tr>
</tbody>
</table>

Contrary to device A, a large fraction of yellow light is “de-trapped” in device B and C. It evidently suggests that successful alleviation of microcavity effect can extract some more yellow emission and enhance EL intensity. This enhancement also agrees with the improved transmission of multilayer cathode. In the transparent device B, a luminance yield of 2.7 cd/A was achieved from the top side but with more than 70% light emitted from ITO anode (bottom side). High reflectance Ag anode of device C forces white light to emit from the top semi-transparent cathode thus leads to very high efficiency of 22.2 cd/A (9.6 lm/W). We believe this significant enhancement in efficiency can be attributed to the combined effect of successful attenuation of the adverse microcavity effect and the improvement of light outcoupling with index matching material [9, 10]. A broad white light emission with a FWHM of 136 nm generated from Device C is also wider than that of bottom-emitting device (116 nm). Although Device C is a broadband-emitting device, it does not have a very good white color. A near white light emission of CIE coordinates of (x = 0.31, y = 0.47) was achieved which is also comparable to that of bottom-emitting device (x = 0.27, y = 0.40). We believe the color of the white device can be further improved by selecting a proper set of emitting materials.

The EL spectra of device C under different viewing angle of 0°, 30° and 60° were shown in Figure 4. In a strong microcavity device, with increasing viewing angle, the peak wavelength shifts rapidly to shorter wavelength and the intensity decreases. It is to be noted that the emission shows less angular dependence in device C as three main peaks of white emission at different viewing angles remain the same.
Current density-Voltage (J-V) and Brightness-Voltage (B-V) curves of these three devices are shown in Figs. 5(a) and 5(b), respectively. It is noteworthy that both J-V and B-V curves of device C show the steepest response. At a high current density of 300 mA/cm², brightness over 57,000 cd/m² was achieved at 10 V which further implies good electron and hole balance in the new top-emitting device was also achieved at the same time.

5. TOLEDs on TFT array

The top-emitting WOLED structure was integrated with a 1.3” QCIF LTPS-TFT substrate. Figure 6 shows the photo images of the panel fabricated with a Cr anode and a Ca/Ag/SnO₂ cathode. It is to be noted that there is no significant color shift of the device at different viewing angle, as shown in Figure 7. The white light emission with CIE coordinates of (0.29, 0.40) was obtained by using three-component emitters to enhance RGB saturation. The simulation after coupled with color filter reveals that the color gamut is increased from 55% to 63%.

6. Conclusion

We have introduced a high-efficiency white top-emitting OLEDs with modified anode and cathode. Removal of ITO layer shortens optical length of the device and alleviates the adverse microcavity effect from which a broad emission of white light is obtained. We believe this highly efficient white top-emitting device may provide a possible solution for the manufacturing of the next generation of high resolution full color LTPS-TFT AMOLED displays.

7. Acknowledgements

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8. References