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Chamika M. Hippola

*Iowa State University and Ames Laboratory*

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OLEDs on Flexible Substrates

Chamika Hippola
Electrical and Computer Engineering
Iowa State University, Ames, Iowa

Introduction

Started as a research curiosity, organic light emitting diodes (OLEDs) have become an essential part in commercial applications. Marking the beginning in 1987 is the fabrication of the first practical thin film fluorescent OLED by C. W Tang et al. at Eastman Kodak. This OLED was based on a thin layer of a small organic molecule, Tris-(8-hydroxyquinoline) aluminum (Alq3), sandwiched between two electrodes; the device operated under 10 V applied bias. Since then, OLEDs have come a long way addressing the need for an advantageous efficient light source in the display industry, with ongoing advances in OLED lighting applications [1]. The innovative development of phosphorescent OLEDs (PhOLEDs) in 1998 by Forrest, Baldo, and Thompson overcame the efficiency restriction faced by fluorescent OLEDs, where the radiative recombination channel is of singlets, which limits the internal quantum efficiency (IQE) to 25% according to spin statistics. With the introduction of a heavy metal atom to the emitting organic compound, such as in platinum octaethylporphyrin (PtOEP), which results in a large spin-orbit coupling, radiative decay of both singlet and largely triplet levels was possible, enabling 100% IQE [2]. Properties such as high brightness and contrast, easy fabrication, and large pixels with large field of view make OLEDs preferable over conventional light sources. Relatively inefficient and short-lived blue OLEDs, with high manufacturing costs, and degradation by water and oxygen are some of the downsides in OLEDs.

OLEDs are typically fabricated on glass substrates, but fabrication on plastic substrates has drawn significant attention for new applications in both lighting and display. The flexible plastic substrates permit roll-to-roll (R2R) processing that enables large scale production, which is cost effective and more efficient than the sheet-to-sheet (S2S) process, where production has been limited by sheet size and transfer delay between process steps [3].

Outcoupling efficiency ($\eta_{\text{out}}$), which is the ratio between the number of photons emitted in the forward hemisphere to the number of photons generated in the OLED, has been low (~17-20%) for conventional OLEDs built on glass substrates. The department of energy (DOE) has set a goal of $\eta_{\text{out}}$ =70% by 2020 [4]. Total internal reflection at the glass-air interface, internally waveguided light in the
organic + anode layers, and dissipation of photons at the metal cathode are the main loss mechanisms in conventional OLEDs. Substrates with a higher refractive index, closer to that of the organics + anode layers, minimize the waveguiding in the latter and can enhance overall light extraction with the addition of e.g., a hemisphere at the air side of the substrate to extract the externally waveguided light (trapped in the substrate). A corrugated substrate minimizes the internally waveguided light, by light scattering and diffraction that result in varying angle of incident at the anode/substrate interface, and possibly also at the substrate/air interface. An external microlens array (μLA) or hemisphere will further extract the light trapped in the substrate, enhancing $\eta_{out}$. Corrugated plastic substrates along with an indium tin oxide (ITO) or PEDOT:PSS anode can be used to build OLEDs with high outcoupling efficiency, as shown in Figure 01.

The figure shows R2R production of an integrated substrate design, which uses transparent polycarbonate (PC) as the substrate. First a nano pattern with suitable features’ pitch and height is imprinted on PC followed by a μLA. Nearly 100% electroluminescence (EL) enhancement was demonstrated using a 2 μm pitch μLA [5]. A transparent external barrier layer needs to be deposited on the μLA to avoid any penetration of water and oxygen that degrade the OLED. A transparent, high-
conductivity conductor, e.g., a fine copper mesh, is formed on the nano pattern, ideally under an ITO anode. This is followed by the OLED deposition using different techniques such as thermal evaporation and inkjet printing. As a final step the device is encapsulated.

Flat and as a result corrugated plastic substrates often present challenges associated with impurities and surface roughness. Sharp points on the substrate’s surface can build very high electric fields that lead to catastrophic shorts. Hence, as a first step toward improving corrugated OLEDs the effect of planarizing the PC substrate was tested by AFM surface roughness measurements and by evaluating the performance of a green PhOLEDs fabricated on planarized PC/ITO. The performance of OLEDs fabricated on planarized PC was compared to that of OLEDs fabricated on as-received substrates. That is, we checked if the PC used in production needs to undergo a planarization process to minimize any surface roughness that is present in the original PC substrate.

**Experimental Details**

Three substrates evaluated in this work were acquired from Microcontinuum Inc. (MCI) as a part of MCI-Iowa State University collaboration. Prior to anode spin coating 93% of poly(3,4-ethylenedioxythiophene) polystyrene sulfonate (PEDOT:PSS), 6% ethylene glycol (EG) and 1% FS-35 surfactant were mixed in a sonicator for 90 minutes. The solution was spin-coated on the PC substrates at 6000 rpm for 30 seconds and annealed for 2 hours. Substrates with the transparent anode were loaded into an evaporation chamber in a glovebox with <15 ppm oxygen. Then 5 nm of Dipyrazino[2,3-f:2’,3’-h ]quinoxaline-2,3,6,7,10,11-hexacarbonitrile (HAT-CN) was deposited by vacuum thermal evaporation. Next 120 nm of 10% of Molybdenum Oxide (MoOx) was co-evaporated with Di-[4-(N,N-dip-tolyl-amino)-phenyl]cyclohexane (TAPC) followed by a 20 nm neat layer of TAPC. This step was followed by evaporating 20 nm of 6% doped Tris[2-phenylpyridinato-C2,N]iridium(III) (Ir(ppy)3) in 1,3-Bis(carbazol-9-yl)benzene (mCP). Afterwards a neat layer of 1,3,5-Tri[(3-pyridyl)-phen-3-yl]benzene (TmPyPb) with a thickness of 20 nm was evaporated. This step was followed by a 40 nm thick 20% Cesium fluoride (CsF) doped in TmPyPb. Finally 1 nm of lithium fluoride (LiF) and 100 nm of aluminum (Al) as the cathode were evaporated. All the thermal vacuum evaporations were performed at a vacuum pressure of 10⁻⁷ mBar.

A Keithley 2400 source meter was used for voltage application and current measurements. The EL was measured at each voltage step using a Minolta LS-110 luminance meter. The EL spectra were
Results and Discussion

To assess the effect of the flat PC, OLEDs on different substrates/anodes were evaluated. First, OLEDs were fabricated on the as-purchased 13 mil (330 µm) thick PC, termed LCC-PC (Device A). Next, the substrate used was the PC planarized by roll-pressing the plastic against a Silicon wafer (Device B). PEDOT:PSS was used as the transparent anode for devices A and B. The third device was fabricated on planarized PC/ITO; the ITO was sputtered at room temperature (Device C). The devices structure was: anode/ HAT-CN (5 nm)/ 10% MoOx: TAPC (120 nm)/ TAPC (20 nm)/ 6% Ir(ppy)3:mCP (20 nm)/ TmPyPb (20 nm)/ 20% CsF: TmPyPb (40 nm)/ LiF (1 nm)/ Al (100 nm).

Figure 02 shows attributes of devices A, B and C. Device C showed the highest brightness of 73,500 Cd m⁻², but this was achieved at a high voltage of ~20 V indicating that the ITO’s conductivity needs to be improved. The turn-on voltage for devices A, B and C were 3.0, 3.1, and 3.8 V, respectively; having a high turn-on voltage confirms the concern of ITO’s resistance. The sheet resistance measured for the ITO on the planarized PC was 130-470 Ω/□, which is significantly higher than the ~12 Ω/□ of commercial ITO on glass. Despite the high turn-on voltage, device C showed superior luminous efficiency and higher external quantum efficiency (EQE) in comparison to the other devices. The luminous efficiencies for devices A, B and C were 50.5, 50.0, and 65.4 Cd A⁻¹, respectively. The respective EQEs were 14.3%, 14.6%, and 18.8%. Atomic force microscope (AFM) was used (performed by Rajiv Kaudal) to study the surface roughness of the PCs and ITO (Table 1). ITO on planarized PC had the largest non-uniform surface roughness of 3.7-5.4 nm Devices A and B presented similar performances with a similar surface roughness, indicating that the planarization is not beneficial and that a smooth, highly conductive is needed for achieving better device performance. The resistance of ITO depends on the sputtering temperature and the layer’s thickness. Increasing the deposition temperature gives rise to larger grains and hence less scattering at the grain boundaries. As a result of the reduced scattering at the grain boundaries, the resistance is significantly reduced [6]. As mentioned, having a rough ITO surface can lead to catastrophic shorts in an operating device and therefore the surface of ITO needs to be smoothened. One way of achieving a lower roughness is by controlling the argon partial pressure in the sputtering system [7]. Reducing the power and increasing the target to substrate distance may also
improve the quality of the ITO. To increase the conductivity, a copper mesh can be embedded into the substrate and ITO sputtered over it, covering the mesh so that sharp edges are avoided.

Figure 02: Performance of devices A, B and C. a) Brightness and current density vs applied voltage, b) luminous efficiency vs brightness, c) power efficiency vs brightness, d) EQE vs brightness, e) EL spectrum and f) photographs of operating device pixels.
Table 01: Attributes of green PhOLEDs on as-received PC/PEDOT:PSS, planarized PC/PEDOT:PSS, and planarized PC/ITO

<table>
<thead>
<tr>
<th></th>
<th>Device A</th>
<th>Device B</th>
<th>Device C</th>
</tr>
</thead>
<tbody>
<tr>
<td>Surface roughness of the substrate (Devices A &amp; B) and of ITO (Device C) (nm)</td>
<td>1.6-5.8</td>
<td>1.5-4</td>
<td>3.7-5.4</td>
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<tr>
<td>Brightness (Cd/m²)</td>
<td>46,180</td>
<td>47,200</td>
<td>73,500</td>
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<tr>
<td>Turn-On Voltage (V)</td>
<td>3.0</td>
<td>3.1</td>
<td>3.8</td>
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<tr>
<td>Luminous Efficiency (Cd/A)</td>
<td>50.5</td>
<td>50</td>
<td>65.4</td>
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<tr>
<td></td>
<td>[14,100]</td>
<td>[6,600]</td>
<td>[5,710]</td>
</tr>
<tr>
<td>Power Efficiency (lm/W)</td>
<td>24</td>
<td>17.6</td>
<td>16.3</td>
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<td></td>
<td>[9,225]</td>
<td>[4,770]</td>
<td>[2,800]</td>
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<tr>
<td>EQE (%)</td>
<td>14.3</td>
<td>14.6</td>
<td>18.8</td>
</tr>
<tr>
<td></td>
<td>[14,100]</td>
<td>[6,600]</td>
<td>[5,710]</td>
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References


