Enhanced Light Extraction from Organic Light Emitting Diodes by Micrometer-Sized Buckles

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The simple ways for creating buckled structures to enhance the light extraction from OLED devices have been investigated. The buckling instability was observed when the ITO was deposited on the polymer-coated glass by sputtering. The textured surface of the ITO layer after buckling was characterized by an atomic force microscopy. The wavelength of the resulting buckled structure was a few microns in a size. The buckling was easily modified by adjusting the pressure of the argon gas during the sputter deposition of ITO layer. The buckled ITO layer was used for fabricating OLED devices. The reduction in the operating voltage for the OLED with the buckled ITO anode was observed. The current and power efficiencies for the OLED with the buckled structure were 5% and 44% higher than those for the conventional OLED. The broader light distribution was observed in the OLED with buckling when the angular dependence of the light intensity was measured.

Keywords: Buckled Structure, Light Extraction, OLED.

1. INTRODUCTION

Due to the applications as displays and solid-state lighting, much effort has been made to improve the performance of organic light emitting diodes (OLEDs) over the last few decades.1–4 However, the lifetime and the external quantum efficiency of OLEDs still need to be improved.5,6 The external quantum efficiency for OLEDs comprises an internal quantum efficiency (IQE) and an extraction efficiency. Theoretically, the IQE reaches 100% by making use of phosphorescent emitters and balancing the charge carriers.7 The extraction efficiency, however, is below 20% because the generated light is trapped at the ITO/glass interface and reflected at the glass/air interface.8 The extraction efficiency for the OLED device has been enhanced utilizing external extraction structures such as hemispherical lens,9 microlens array,10 antireflective surfaces,11 and scattering layers12 on the outside of the substrate. This approach, however, is less efficient because only the light trapped by the substrate mode can leave the device. Another approach to enhance the efficiency of OLED is incorporating internal extraction structures between the substrate and the anode, which can extract the light otherwise trapped by the waveguided mode.13–15 Introducing the particles with the high refractive index into the space between the anode and the substrate not only scatter the light but also reduce the reflection at the interface of two layers. Recently, Chang group reported that the OLEDs with the scattering layer of TiO2 nanoparticles enhanced the power efficiency by more than 400%.16 Do group has reported that the current efficiency for the OLED with photonic crystal structures as the internal extraction structures was higher than the conventional OLED by 85%.17 However, the angular dependence of the emitted color was observed. As the angular-dependent colors are not suitable for OLED luminaires, novel internal extraction structures have been suggested to enhance the efficiency of the OLED.6 In 2010, Koo et al. reported that the OLEDs incorporating random scattering structures between the glass substrate and the indium tin oxide (ITO) electrode enhanced the efficiency by 90% with no change in the emission spectrum compared to the control device.18 The random scattering structures were formed through the spontaneous buckling of the aluminum layer on the PDMS layer utilizing the different thermal
expansion coefficient between two layers.\textsuperscript{19} Inspired by this report, Ju and coworkers have fabricated the OLEDs with textured surfaces which enhanced the power efficiency by 102%.\textsuperscript{20} The textured structures were formed by etching the quartz substrate with CHF\textsubscript{3} employing a poly(methyl methacrylate) as a mask. The patterns formed were duplicated on the polymer-coated substrate supporting the OLED stack through nanoimprint lithography later. However, the nanoimprint lithography was not applicable for a large area because of the pattern collapse.\textsuperscript{21} Here, we reported that the spontaneous buckling of a polymer on a glass substrate during
sputtering deposition of ITO. In this process, the nanoimprint lithography was not required. The luminous efficacy for the OLED device with the buckled structure was improved by 44% compared to that for the OLED without buckling.

2. EXPERIMENTAL DETAILS
2.1. Materials
The photoresist (THB-151n) was purchased from JSR Micro. Methyl alcohol, isopropyl alcohol and ethyl acetate were purchased from Aldrich.

2.2. Preparation of the Buckled ITO Layer
The bare glass was cleaned with water, methyl alcohol and isopropyl alcohol. The concentrated photoresist was diluted with ethyl acetate to afford the diluted photoresist with solid content of 70%. The concentrated and diluted photoresists were spin-coated on the bare glass, pre-baked at 90 °C for 1 min and irradiated with UV radiation for 10 seconds to afford the polymer-coated glass substrates. The thickness of the polymer film was measured on an Alpha-Step P-10 Profilometer (KLA-Tencor corporation). The indium tin oxide (ITO) layer with the thickness of 150 nm was deposited on the as-prepared polymer-coated glass using the RF sputtering system equipped with the ITO target at power of 100 W in argon atmosphere. The morphology of the ITO surface was characterized by an atomic force microscopy. The thickness of the ITO film was measured on an Alpha-Step P-10 Profilometer (KLA-Tencor corporation). The sheet resistance of the ITO layer was measured by a four point probe (Lorestra-GP (MCP-T610, Mitsubishi chemical analytech co., LTD.).

2.3. Fabrication of OLED Devices
In this study, we have fabricated two types OLED devices which consisted of a \( N,N'\)-Di-[(1-naphthyl)-\( N,N'\)-diphenyl]-1,1'-biphenyl)-4,4'-diamine (NPB, 600 Å) film as a hole transport layer, an Ir(ppy)\(_3\) doped 4,4'-Bis(N-carbazolyl)-1,1'-biphenyl (CBP, 320 Å) film as an emitting layer, a 1,3,5-Tris(1-phenyl-1H-benzimidazol-2-yl)benzene (TPBi, 200 Å) film as an electron transport layer, a LiF (5 Å) film as an electron injection layer, and an Al (1200 Å) film as a cathode. In a vacuum chamber (\( \sim 10^{-6} \) Torr), the organic materials and aluminum were thermally deposited at a rate of \( \sim 2 \) and \( \sim 4 \) Å/s, respectively. The active area of the device was \( 4 \times 6 \) mm\(^2\).

2.4. Characterization of the Device Performance
The current–voltage–luminance characteristics of the OLEDs were measured on a source measurement unit (Model 2400, Keithley Instruments, Inc.) and a spectroradiometer (CS-1000, Konica Minolta, Inc.). An angular-dependent emission of a device was measured by attaching an OLED on a stage which rotates from 0° to +85° in a step of 5 degree. A power efficiency of a device was measured using an integrating sphere equipped with a source measurement unit (Model 2425, Keithley Instruments, Inc.) as a power source.

3. RESULTS AND DISCUSSION
The buckled pattern was formed during the ITO deposition on the polymer-coated glass substrate using RF sputtering.

![Figure 3](image-url)
Enhanced Light Extraction from Organic Light Emitting Diodes by Micrometer-Sized Buckles

Kim et al.

Figure 4. The current density–voltage–luminance characteristics of the device A with buckling (●) and device B without buckling (▲). (a) Current density versus voltage of the devices. (b) Luminance versus voltage of the devices. (c) Current efficiency as a function of voltage of the devices. (d) Normalized electroluminescence spectra of the devices.

(Fig. 1). The argon plasma produced in the sputter chamber affected buckling of the polymers. To elucidate the plasma effect on the buckling, the argon gas pressure varied from 2 mTorr to 7 mTorr during the ITO sputter deposition (Fig. 1). The resulting morphology of the ITO-coated glass was characterized by an atomic force microscopy (AFM). At 2 mTorr, the wavelength and the amplitude of the buckled structure were ca. 5 μm and ca. 1 μm, respectively. As increasing the pressure of argon from 2 mTorr to 5 mTorr, the wavelength and the amplitude was little changed in the pattern. In the range between 2 mTorr and 5 mTorr, the ITO-coated glass was translucent owing to the buckled structure with greater than 700 nm in amplitude. The ITO-coated glass obtained at 7 mTorr pressure of argon, however, was almost transparent in appearance. The AFM image revealed that the amplitude of the buckled structure was smaller than 20 nm. It should be noted that the pressure also affect the uniformity of the deposited ITO film on the polymer-coated glass. In our experiment, the increased pressure of argon resulted in the different thickness of the deposited ITO film over the entire glass substrate due to the high depositing speed. The thickness of the deposited ITO film on the glass with 5 × 5 cm² area was uniform when the argon pressure was 3 mTorr. The pressure of 2 mTorr required the extended time to deposit the ITO film. The sheet resistance of the ITO film obtained at 3 mTorr was ∼50 Ω/□.

Figure 5. (a) The photographe of the anode surface of the OLED device with buckling. A few micron-sized buckled structures were observed using a microscope. The buckling was induced when the ITO layer was deposited on the polymer (7 um) at the Ar pressure of 3 mTorr. (b) Angular dependence of normalized light intensity of the device A with buckling (dashed line) and the device B without buckling (solid line). The light distribution was broader in device A due to the buckled structures. (c) The angular dependence of CIE color coordinate emission in device A with buckling. The color change ratios were Δx = 4.2% and Δy = 3.7% as increasing the viewing angle from 0° to 85°.
The concentrated photoresist which was spin-coated on a glass at a speed of 5000 rpm afforded the thickness of 11 μm after the UV curing process due to its high viscosity. In order to reduce the thickness of the polymer layer, the solution with a solid content of 70–90% by weight was prepared by diluting the concentrated photoresist with ethyl acetate. When the solid content was less than 70%, the solution was not suitable for a spin-coating process due to its low viscosity. The thickness of the polymer layer was reduced from 8 μm to 7 μm as the solid content reduced from 90% to 70%. The AFM image revealed that the buckled patterns of the ITO layers were similar regardless of the concentration of the photoresist solution (Fig. 2). For the buckled anode, the ITO film was deposited on the 7 μm polymer layer at 3 mTorr argon using RF-sputtering.

Encouraged by this, OLED devices with and without the buckled structure have been fabricated. The ITO-coated glass without the buckled structure was produced by depositing ITO film on a bare glass. To minimize the variations, the ITO-coated glasses with and without buckling were prepared at the same batch and used for OLED fabrication. The device A and B consist of ITO(with buckling)/NPB/CBP:Ir(ppy)3/TPBi/LiF/Al and ITO (without buckling)/NPB/CBP:Ir(ppy)3/TPBi/LiF/Al, respectively. Interestingly, the uneven aluminum layer in device A was observed by naked eye after fabricating the OLED through thermal evaporation in the vacuum chamber. The AFM image of the aluminum layer displayed the buckled structure existed and it was dissimilar with the buckled patterns of the ITO layer and the polymer layer (Figs. 2 and 3). The buckled cathode indicated that the corrugated structure was formed after the organic layer and Al was sequentially deposited on the buckled anode (Fig. 3).

The I–V–L characteristics of the devices were measured until a voltage reached 8 V (Fig. 4(a)). The current-voltage curves showed that the current density was higher in device A than B due to the buckling. The higher luminance value was observed in device A than device B (Fig. 4(b)). The increased luminance was attributed to the increase in emitting area by the buckling in device A. At a brightness of 1000 cd/m², the current efficiency of device A and B was 13.0 cd/A and 12.4 cd/A, respectively. The current efficiency of device A was 5% higher than that of device B. Both emission spectra of device A and B featured the maximum intensity at 515 nm indicating that the randomness of the buckled structure in device A led no change in color (Fig. 4(d)).

The buckling patterns with a few micrometers in size were observed by an optical microscope while the device A was turned on (Fig. 5). In order to investigate the effect of the buckling on the light distribution, the angular dependence of the scattering intensity was measured. As a result, the broader distribution of the emission was observed in the device A (Fig. 5). Due to the different emission distribution between the device A and the device B, the extracted light was carefully measured as a function of an electric current using an integrating sphere. The power efficiency at 5 mA was 7.5 lm/W for the device A and 5.2 lm/W for the device B suggesting that the enhancement factor was 1.44. The higher enhancement in power efficiency than in current efficiency was attributed to the decreased operating voltage as well as the broader light distribution in the corrugated structure. Last but not least, there was little color change when the angular-dependent color of device A was measured utilizing Commission Internationale de’Eclairage (CIE) color coordinates due to the directional randomness of the buckled structures (Fig. 5(c)).

4. CONCLUSION

In sum, the spontaneous buckling was investigated by depositing ITO film on the polymer-coated glass. The buckling was affected by the pressure of argon during sputtering deposition of ITO layer. The OLED with the buckled ITO layer enhanced the power efficiency by 44% compared to the conventional OLED.

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References and Notes


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