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Efficiency enhancement of organic light-emitting devices by using honeycomb metallic electrodes and two-dimensional photonic crystal arrays

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ABSTRACT

In this paper, a facile fabrication technique called nanosphere lithography combining with two-step reactive ion etching method for patterning honeycomb metallic electrode with high transparency and excellent uniformity is demonstrated. The patterning silver electrode with 15-nm film thickness and 68.6% fill-factor was used as the organic light-emitting diode (OLED) anode, which showed an average transmittance of 77.4% and sheet resistance of 30.7 Ω/□. The current efficiency is 8.35 cd/A for the OLED with patterned silver anode under 100 cd/m^2 operation brightness, which was 47% higher than the device with indium tin oxide (ITO) anode. After applying the polystyrene nanosphere to form a photonic crystal array onto the device, the extracted light from organic mode can be further coupled out from device substrate mode. The overall luminous enhancement of the device with the combination of internal honeycomb metallic anode and external photonic crystal array is 115% higher than the traditional ITO-based OLED.

1. Introduction

Organic light-emitting diodes (OLED) have been studied intensively for their numerous applications in the area of flat panel display and general illumination due to the promising advantages of excellent electroluminescence performances, good flexibility, light weight, better durability, and low power consumption, and so on [1–3]. However, due to the mismatch of refractive index, there is only a small fraction of the generated light in organic layers can escape form the device due to the total internal reflection (TIR) at the glass/air interface and wave-guiding effect in the high-index organic layer [4,5]. Many researches have been reported that the light extraction efficiency (escaped photons/generated photons) of OLEDs is less than 30% [6,7]. Numerous approaches have been proposed to reduce the wave-guiding mode of the organic layers, such as introducing textured surface, insertion of low-index materials and fabricating patterned nanostructures into the device [8–11]. But the light extracted from organic layers also suffers from the light confinement due to the TIR between glass substrate and air. By applying light-coupling structures onto the surface of device can further extract confined light [5,12,13]. In our previous study, higher luminous improvement of device with internal light extraction nanostructures has been achieved by

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attaching microlens array (MLA) on the substrate surface. The microlens array was fabricated by using photoresist thermal reflow method and a double replica molding process [14].

On the other hand, indium tin oxide (ITO) is the most widely used material as the transparent conductive electrode. Owing to the limited supply and high cost of indium in recent years, many studies have proposed different materials to replace the ITO electrode, such as conductive polymers, carbon nanotubes, and conducting oxides [15–18]. In addition to those proposed materials, patterned metallic electrode with good conductivity and high transparency can act as an excellent transparent anode alternative [12,19]. For patterning an aligned nanostructure arrays, photolithography techniques such as atomic force microscopy (AFM) lithography, electron-beam lithography (EBL), laser interference lithography (LIL), and scanning tunneling microscopy (STM) lithography were widely applied for the formation of micro/nanoscale periodic patterns [20–24]. But above methods are complicated and need high fabrication cost. Van Duyne et al. firstly proposed a facile and effective nanofabrication method called nanosphere lithography (NSL) for the production of periodic particle arrays with nanometer scale features [25,26]. In the simplest NSL scenario, only a monolayer of hexagonally close-packed nanospheres is self-assembled onto the substrate. When one deposits metal through the monolayer mask, the three-fold interstices allow deposited metal to reach the substrate, creating an array of triangularly shaped nanoparticles with P_{6mm} symmetry. Over the past decade, NSL has been widely employed in fabricating two- and three-dimensional (2- and 3D) micro/nano structures, such as quantum dots, nanowires, nanomesh, antireflection structure (ARS), and 3D inverse-opal photonic crystals [19,27–31]. Compared to those traditional lithographic methods, NSL is not only a simple but also a cost-effective route for the patterning of long-range periodic nanostructure arrays in a large scale.

In this paper, a honeycomb metallic electrode with 15-nm silver film thickness and 68.6% fill-factor (ratio of hole area to total area) made by the NSL is acted as the transparent anode. The periodic metallic nanohole arrays can effectively enhance the light extraction efficiency of OLEDs. The current efficiency is 8.35 cd/A for OLED device patterned with Ag anode under 100 cd/m^2 operation brightness, which has 47% improvement higher than the device combined with traditional ITO anode due to the better light extraction. In addition, the trapped photons in the glass substrate were further extracted to the air by attaching a photonic crystal layer made of polystyrene (PS) nanosphere array onto the device surface. The total luminous enhancement for the device patterned with silver electrode combining with the photonic crystal layer shows a great efficiency improvement of 115% as compared to the traditional ITO-based OLEDs.

2. Experimental procedure

The schematic diagrams of fabrication process for the OLED devices with patterned silver anode were shown in Fig. 1. The whole process can be divided into two major parts, including the patterning of metallic electrode via nanosphere lithography and evaporation of conventional organic layers. In the part of nanosphere lithography, the surfactant-free monodispersed PS nanosphere suspension (10 wt% in water with mean diameter of 590 nm) was purchased from Bangs Laboratories Inc. and diluted by methanol solution containing a surfactant Triton X-100 (400:1 in volume) by a factor of 2.0 in ultrasonic bath. First, the as-prepared diluted latex PS nanosphere solution was spun-coated onto a clean glass substrate as a mask of hexagonal array. Prior to the spin-coating process, the glass substrate was thoroughly cleaned with sulfuric acid (H_2SO_4) mixed with hydrogen peroxide (H_2O_2) and then rinsed by deionized water to achieve a hydrophobic glass surface to help the followed NSL process to form a monolayer hexagonal close-packed nanosphere arrangement. The period of the hexagonal arrays was 590 nm which is determined by the initial diameter of PS beads. To further reduce the diameter of 590-nm PS nanospheres, we used a home-made reactive ion etcher (RIE) to etch the edge of the PS nanospheres by two-step oxygen plasma etching method to precisely control their size and adjust the fill-factor. Subsequently, the air-gap between the nanospheres was filled-up with silver thin films by electron-gun evaporation. Finally, those as-prepared samples were put into the dichloromethane (CH_2Cl_2) solution in an ultrasonic cleaner for 15 min. After the lift-off process, the PS nanospheres were removed and then the periodic honeycomb silver thin film was successfully formed on the flat glass substrates.

Fig. 2(a) shows the layer structures for the OLED device. The OLED device was made on the honeycomb silver anode with surface treatment by the self-assembled monolayer (SAM) of binary mixtures of n-decanethiol and fluorinated analogue. It was not only acted as a protection layer to smooth the patterned silver surface but also increased the work function of silver films from 4.68 to 5.6 eV, providing a better energy level alignment between the anode and the HOMO level of the NPB as shown in Fig. 2(b). After the SAM surface smoothing treatment, organic layers were deposited in an ultra-high vacuum thermal evaporation (around 5×10^{-6} torr). Hole-transporting layer (NPB), red fluorescent layer (Alq_3:1%DCJTB), and electron-transporting layer (Alq_3) were sequentially evaporated onto the patterned silver anode. Fig. 2(c) shows the molecule structures of NPB, Alq_3 and DCJTB, respectively. Then LiF and Al were set as the opaque cathode for the OLED device. The detailed device structure is honeycomb Ag anode/NPB(50 nm)/Alq_3:DCJTB(40 nm)/Alq_3(45 nm)/LiF(1 nm)/Al (100 nm). The Alq3 doped with 1% DCJTB as fluorescent layer is the typical control device for red OLEDs. Some research has shown that the DCJTB concentration significantly affects the electron mobility in Alq3:DCJTB [32]. For relatively low doping concentration (<1%), the electron mobility of Alq3:DCJTB decreases with the doping level. The mobility is then increasing if the dopant concentration is further increased, followed by a decrease for doping levels larger than 2%. Also, from the work represented by Y.T. Chang et al., Alq3 doped with 1% DCJTB shows high quantum yield of around 90% and color saturation with a