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Random nano-structures as light extraction functionals for organic light-emitting diode applications

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ABSTRACT

In this study, we demonstrated a nano-structured random scattering layer (RSL) as an internal light extraction method to improve the light extraction efficiency of organic light-emitting diodes (OLEDs). Using dewetted Ag droplets as a hard mask, we textured the glass surface to have a scattering layer of the random structure. OLEDs equipped with the RSL showed more that 50% improvement in the external quantum efficiency (EQE) and luminance efficacy (LE) compared to OLEDs without the RSL. This improvement can be understood by the scattering effect which reduces the optical loss at wave-guided modes. Also, by combining the RSL and an external light extraction micro-lens array (MLA), it was possible to achieve further improvements of 105.8% and 92.06% in the EQE and the LE, respectively.

1. Introduction

In organic light-emitting diodes (OLEDs), injected electrons and holes recombine in the organic emissive layer to generate electroluminescent light. A great advancement in the synthesis of organics and energy level designed electrodes has enabled the achievement of 100% internal quantum efficiency [1–5]. In stark contrast, most OLEDs suffer from poor external efficiency of 20% and even lower. This is explained by the fact that the outward traveling light is confined due to the difference in refractive indices of the constitutive layers of the device [6–9]. The efficiency limit posed by the optical problems is particularly detrimental in the field of OLED lighting, in which a large-area illumination surface is favored. To increase the luminous intensity, higher power must be applied. This not only increases the power consumption but also deteriorates the emissive organics. Conversely, overcoming the external efficiency problems can result in improved energy saving and extended lifetime of OLEDs. To improve the light extraction efficiency, various technologies for internal and external substrate modification have been proposed [10–15]. Most methods rely on rather complex processes, which might not be readily applied in practice. Also, most reported light extraction methods have been hampered by low transparency of the light extraction inserts. By recasting the internal surface, which faces the organic layer of the glass substrate into a layer with random texture, scattering effects can be achieved, which can contribute to improving the light extraction efficiency. The layer with random texture alters the incidence angles of light rays from the light emitting part to the glass substrate, resulting in a reduced waveguide mode [16–19]. By using random texture, the problem of enhancement or reduction of particular wavelengths can be avoided. Such a feature is very important in general lighting, in which white light is widely used. In this work, as an effort to improve the light extraction efficiency of OLEDs, we have reshaped the surface of the glass substrate to have a scattering layer of

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random nano-structure. The presence of three dimensional features can alter the traveling path of the light. Once the feature size becomes comparable to the visible light wavelength, the traveling light may experience the dispersion and redirection in the forward direction. Collectively, these phenomena are called the scattering, which can be effectively applied to OLED light extraction technology. Our light extraction approach can be summarized as following. We have placed a light extraction layer, which is in touch of the interior surface of the glass surface. The light extraction layer is composed of a high refractive index (n) matrix and a collection of randomly dispersed scattering features, which embedded in the matrix. In this structure, first, the generated light is extracted from the indium tin oxide (ITO) layer using a material of high n. And then, the light scatters at the embedded three dimensional features. This approach can significantly reduce the loss, which is due to the difference in refractive indices between ITO (nITO $\sim$ 1.9) and glass (nGlass $\sim$ 1.5), and result in enhanced light extraction efficiency. The major technical difficulty is the formation of scattering features, which have comparable sizes of the visible wavelengths. Various technologies have been suggested to fabricate nano-structures, including electron-beam or nano-imprint lithography, colloidal lithography, nano-sphere formation, and template methods [20–24]. However, more scalable and cost effective methods are still needed to be applied to OLED productions. In this work, we have used spontaneously dewetted Ag droplets as the hard mask to obtain randomly distributed scattering features. The thermally assisted dewetting process has the advantage of being spontaneous; therefore, it can be cost effective and scalable. Masks were used to dry etch the glass surface into a scattering layer of random texture. Because the glass itself is textured directly, the random scattering layer (RSL) is structurally continuous to its support and does not require any additional optical interfaces. OLEDs equipped with RSLs showed improved external quantum efficiency (EQE) and luminance efficacy (LE). The use of dewetted metallic films to form the RSL is facile and practical for enhancing the performance of OLEDs.

2. Experiments

Fig. 1 shows the process to form the RSL. On the glass substrate, an SiO$_x$ layer of 500 nm thickness and an Ag film of 60 nm thickness were deposited sequentially (Fig. 1a). SiO$_x$ and Ag were deposited by plasma-enhanced chemical vapor deposition and thermal evaporation methods, respectively. To form the Ag mask, the sample was heated to 400 °C in order to rupture the Ag film; this process resulted in irregularly distributed Ag particles. Nano-sized Ag particles were formed by dewetting process. To form the RSL, the exposed portion of the SiO$_x$ layer was etched using an induced coupled plasma etching method. A mixture of CF$_4$ and Ar gas was used as the etching gas. Later, the Ag mask was removed using HNO$_3$ etchant. The RSL consists of irregular nano-sized islands with random distribution. Because positional correlation is absent among the islands, the problem of optical wave guiding can be avoided. If wave guiding is present, the spectra of white light, which is the color for general lighting, can be distorted. On the RSL, a planarization layer was coated. Planarization is necessary to stabilize the forthcoming deposition of organics and also to suppress any anomalies in the current flow. Our phosphorescent OLEDs consisted of a stack structure of ITO (100 nm)/N,N°-diphenyl-N,N°-bis(1-naphthyl)-1,1°-biphenyl-4,4/-diamine [NPB] (30 nm)/1,4,5,8,9,11-hexaazatriphenylene hexacarbonitrile [HAT-CN] (10 nm)/NPB (30 nm)/HAT-CN (10 nm)/NPB (30 nm)/HAT-CN(10 nm)/NPB (35 nm)/tris(4-carbazoyl-9-ylphenyl)amine [TCTA] (5 nm)/Phosphorescent emission layer (10 nm)/2,6-bis-[3-(carbazol-9-yl)phenyl]pyridine [DCzPPy] (10 nm)/1,3-bis(3,5-dipyrid-3-yl-phenyl)benzene [BmPyPB] (60 nm)/LiF (1 nm)/Al (100 nm). The emission layer gives a green color, with its main peak at $\lambda = 540$ nm. The RSL infrequently contains protruding features, which may cause electrical instability during the device operation. In order to stabilize the device, we used a hole-transport layer (HTL) with a thickness of 160 nm. Using a thick HTL may increase the operating voltage. To suppress the increase, we used an NPB and HAT-CN alternating HTL layer. Such a stacking sequence enhances the hole transport toward the emission layer [25,26]. The active luminous area was 7×10 mm^2. All materials were electronics grade and were used without further purification. All organic layers were deposited in a high vacuum chamber below 6.67×10^{-5} Pa using a thermal evaporation method. ITO and LiF/Al were the anode and the cathode, respectively. To protect the organics from atmospheric degradation, the fabricated OLEDs were glass encapsulated in a glove box.

The current density–voltage (J–V) characteristics were measured using a current/voltage source unit (Keithley 238). The angular-dependent luminance and electroluminescence (EL) spectra were measured with a goniometer-equipped spectroradiometer (Minolta CS-2000), at a constant current density level of 1.5 mA/cm^2. The transmittance of the RSL was measured using a spectrophotometer (Hitachi U-3501).

3. Results and discussions

3.1. The random scattering layer

Fig. 2 shows scanning electron microscopy (SEM) images of the surface morphology of the SiO$_x$ layer (a), the Ag mask irregular distribution (b), the RSL (c), and the planarization layer on the RSL (d). The dewetting of the Ag films is a response of the system to minimize the total energy of the system [27,28]. Qualitatively, Ag dewetting indicates that the surface energy of Ag is higher than that of the support. Dewetting can be facilitated via an additional process of thermal annealing. Theoretically, there are two different mechanisms for the dewetting process, namely, spinodal dewetting and nucleated dewetting [29]. Spinodal dewetting occurs when thermal action amplifies the surface capillary wave or perturbation to dewet the films. Because the entire film is perturbed, the dewetting process is homogenous and usually yields