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Polymer-based scattering layers for internal light extraction from organic light emitting diodes

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ABSTRACT

Efficient light extraction for organic light emitting diodes (OLED) using scalable processes and low-cost materials are important prerequisites for the future commercialization of OLED lighting devices. The light-extraction technology exhibited in this paper uses polymer-based high-refractive index scattering layers processed from solution. The scatter matrix formulation incorporates two types of nanoparticles for refractive index tuning and scattering, respectively. Planarization by the same material in order to reduce surface defects was critical for achieving highly increased device yield. Highly efficient and defect-free large-area (1.8 cm^2) white OLED devices were fabricated on top of the scattering layer in a bottom emitter configuration. Light extraction enhancement leads to an overall efficiency gain of up to 81% for luminances of 5000 cd m^{-2}.

1. Introduction

Today, organic light-emitting diodes (OLEDs) are a promising technology for the future of illumination. OLED's unique features like transparency, flexibility and thinness offer new possibilities for design and application. However, to be competitive with conventional light sources, the price per lumen needs to be reduced [1]. Increasing light extraction is an appropriate approach to increase the performance of the device [2]. However, the additional cost of light extraction measures should be kept to a minimum. Many schemes for increasing the light extraction in OLEDs have been proposed [3]. In this work, we present an outcoupling technology for internal light extraction which combines the benefits of low-cost materials, scalable processes, and high efficiency gain.

Due to the different refractive indices of the substrate ($n \sim 1.5$) and the organic layers ($n \sim 1.8$) — where light emission takes place — light is trapped by total internal reflection. The organic layers are sandwiched between a reflective metallic cathode and a transparent anode (indium tin oxide - ITO). Around 30% is trapped in the glass substrate and ~50% in the high refractive index region of the ITO/organic layers [4]. Hence, only ~20% of the generated light can escape the device. The angular range in that the escaping light propagates is called the escape cone. By adding a light extraction layer, the trapped light can be redistributed, so that some of it also reaches the escape cone.

The light trapped in the substrate can be accessed using external extraction on the side of the substrate that faces air. Common techniques are micro lens arrays or scattering layers attached to the substrate after processing the OLED devices on the opposite site of the substrate [5,6] The substrate modes and the ITO/organic-modes can be redistributed by introducing internal light extraction layers (IEL), which results in enhanced light out-coupling [3,7,8]. To access the light trapped inside the organic layers, the refractive index of the IEL layers needs to be equal to or higher than that of the ITO/organic layers. Equally important for the efficiency gain induced by the internal light extraction layer are the layer's other optical properties such as haze and absorption. The lower the absorption of the IEL, the higher the overall efficiency.

Light scattering, micro lenses or photonic structures can be used for internal light recycling [3]. Some of those techniques are either very expensive or cannot be applied to large areas or high volume

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throughput. A promising low-cost approach with commercial potential is the use of volume scattering layers. Shiang et al. already presented a deep discussion about the outcoupling of light for volumetric scattering layers of different haze values [9]. C.-H. Shin et al. reported scattering layers made of polystyrene with embedded Al2O3 particles to enhance the light extraction of OLEDs by 40% [10]. The absolute values of the external quantum efficiency (EQE) were conducted at a current density of 53 mA cm^{-2} and are raised from 1.2% to 1.8%. For further improvement an index-matched scattering medium is needed, to access and redistribute wave-guided ITO/organic modes. Chang et al. reported a fourfold power efficiency improvement at 5000 cd m^{-2} of blue phosphorescent OLED by using nanocomposite scattering layers with high refractive index, which consist of a transparent photoresist and TiO2 particles in two different sizes [11]. Small particles with a size of 25 nm were used to raise the refractive index and larger particles to induce scattering. Blue-emitting OLED devices were manufactured on top of the scattering layers with an active area of 2×2 mm^2. The external quantum efficiency (power efficacy) of those devices at 1000 cd m^{-2} were raised to 25.2% (37.1 lm W^{-1}) compared to 11.0% (11.6 lm W^{-1}) of the reference with a strong roll-off for higher luminances. The resulting gain factors showed a strong variation with current density. The results are promising but do not show if the technology is applicable on large areas with a high production yield. In the present work, a similar nanocomposite material system is evaluated and the results are shown on larger areas, with a statistic of plenty manufactured white-light emitting OLED devices to show the need for planarization layers on those composite scattering layers, in order to achieve high production yield.

Besides refractive index matching of the IEL, the surface roughness and especially surface defects are critical properties [12–14]. Both holes and spikes can result in electrical defects or short circuit the OLED devices deposited on the IEL. Each electrical defect leads to a device failure. For general illumination, large emitting areas are needed, and thus the surface of the IEL layer must be defect-free on a large scale. Most of the current publications evaluate IEL layers on very small OLED areas, and so if considered for the large areas required for commercialization, many of the presented techniques are too complex, hard to manufacture, very expensive, or not feasible for high volume throughput. This work consequently concentrates on low-cost materials and processing and exhibits the potential for future application to large area OLED devices.

2. Results and discussion

2.1. Analysis of the scattering layers

A scattering layer needs to fulfill several requirements in order to be used as an internal light extraction layer in OLED devices. The requirements for the particular device architecture, shown in Fig. 1, are processing-, surface-quality- and optics-related. Solution processing is a fast and scalable approach for layer deposition. Many different techniques are possible, including offset or inkjet printing, slot die coating or doctor blading.

An optical analysis of the two scattering layers used as IEL in device type I and II is shown in Fig. 2 a). This angular resolved scattering measurement shows similar light distributions for both layer architectures, which indicates that the additional planarization coating, stacked on top of the scattering layer, does not affect the scattering capabilities. Direct transmission and specular reflection for the planarized layer is slightly higher, due to reduced surface roughness and therefore suppressed surface scattering. The resulting haze induced by the IEL is ~0.75 (±0.05) evaluated at a wavelength of 550 nm. The refractive index of the matrix of the volume scattering layer, shown in Fig. 2 b), is higher or at least equal to that of the organic layers (n ~ 1.7–1.9) and thus light extraction of both ITO/organic-modes and substrate modes is expected. The tailored refractive index of the material can be described by the effective medium approach and can be calculated according to the volume fractions of the polymer and the nano-fillers [15]. The extinction coefficient, also shown in Fig. 2 b), is below $<10^{-3}$ in the region of the typical OLED emission spectrum, between 450 and 700 nm, which is sufficiently low in order to neglect absorption losses. Beside optical properties, such as absorption within the scattering layer by either the polymer matrix or scattering particles, the surface quality of the IEL is a key requirement for zero-defect OLED deposition. This is the most critical attribute when applying the particle-based polymeric scattering layer to OLEDs. Due to the architecture, all of the active layers were deposited on top of the scattering layer. Each topological defect can lead to an electrical defect in the OLED devices. Both spikes and holes in the surface of the IEL represent critical defects for further OLED processing. To reduce roughness, spikes and holes, a planarization layer is introduced on top of the IEL. It is made of the same high refractive index material used for the matrix of the scattering layer, but without the dispersed scatter particles. Therefore it is perfectly index-matched to the IEL and has no effect on light extraction capabilities, neglecting general absorption.

To evaluate the surface quality of the IEL and the planarized IEL respectively, Atomic Force Microscope (AFM) and Scanning Electron Microscope (SEM) measurements were obtained. The AFM and SEM images of the IEL and the planarized IEL are shown in Fig. 3. The overall roughness is dramatically reduced by the planarization layer. For a 20×20 μm^2 area measured by AFM, the root-mean-square roughness was reduced from 35 nm to 7 nm. In other words, planarization reduces roughness by a factor of 5. It should be noted that the roughness was only measured on a small area and is therefore merely an indicator that the layer may be usable for further OLED deposition. For a complete validation of the surface quality, the OLED device itself is the best test vehicle, as surface irregularities will result in shunts and easily detectable optical artifacts.

2.2. Analysis of OLED devices

White-emitting OLED devices, with an active area of 1.8 cm^2, were fabricated on the pure (device type I) and planarized (device type II) IEL. Devices with the same stack and size, without any additional layer between the glass substrate and the active stack, were fabricated as a reference. An overview of the used device structures is shown in Fig. 1. A tandem structure was used [16], with a phosphorescent yellow emission unit (Y unit) and a fluorescent blue (B unit) one separated by a charge generation layer (CGL) [17,18]. Electron and hole injection layers (EIL/HIL) and electron and hole transport layers (ETL/HTL) inject charge carriers into the device and transport them to the emission units [19]. The total thickness of the organic stack was 509 nm. The layer architecture was optimized by simulation to achieve white emission according to the simulation method presented by D. Setz et al. [20] The optimization was performed for maximum light output to air at a targeted white color point. It should be noted that our results are not specific to the details of the material set used and similar results can be achieved using other OLED stack architectures. For statistics, 21 devices each of types I and II were manufactured and seven of the reference.

All devices were electro-optically characterized. The current–voltage characteristics of the three device types are shown in Fig. 4 a). All devices exhibit a characteristic diode behavior with