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Letter

Improved light outcoupling efficiency in organic light-emitting diodes with nanoparticle-embedded charge transport layers

Jin Hee Kim, Joo Won Han, Dong Jin Lee, Siti Aisyah Nurmaulia Entifar, Zeno Rizqi Ramadhan, Kwon Taek Lim, Yong Hyun Kim[*]

Department of Display Engineering, Pukyong National University, Busan 48513, Republic of Korea

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ABSTRACT

We demonstrate high performance internal light outcoupling structures using nanoparicle-embedded hole transport layers (NE-HTLs) for solution-processed organic light-emitting diodes (OLEDs). The NE-HTLs show smoothly formed corrugated surfaces. The OLEDs with NE-HTLs show enhanced power efficiencies by a factor of 1.44 for polystyrene (PS) nanoparticles and 1.78 for SiO_2 nanoparticles, respectively. These improvements are mainly attributed to the reduced total internal reflection at the interfaces of organic layers and transparent electrodes in devices by introducing NE-HTLs. Because the internal light outcoupling structures are prepared on top of the transparent electrode, a damage of organic layers and deterioration of the electrical and optical properties of transparent electrodes are not induced. In addition, the OLEDs with NE-HTLs show the angle-independent light outcoupling enhancement. It is expected that the NE-HTLs fabricated by the simple solution process can contribute to the development of efficient internal outcoupling structures for OLEDs.

1. Introduction

Organic light-emitting diodes (OLEDs) have shown great promise in efficient display and next-generation lighting applications because of their desirable features such as low power consumption, steadily improving efficiency, excellent mechanical flexibility, and high color purity [1,2]. These special properties of OLEDs can get a large share of the global lighting markets together with conventional light emitting diodes (LEDs). While an internal quantum efficiency of OLEDs already approaches 100% with the use of phosphorescent emitter materials, the external quantum efficiency of typical OLEDs is significantly limited by the low light outcoupling efficiency resulting from the total internal reflection at the interfaces of optically distinctive layers [3,4]. The light outcoupling efficiency of typical planar bottom-emitting OLEDs is limited to only about 20–25% due to the waveguide mode at the interface of indium tin oxide (ITO)/organic layers, the substrate mode at the interface of air/substrate, and surface plasmon polaritons (SPPs) at the metal electrode [3,5].

Accordingly, many approaches have been proposed to extract light trapped in substrate mode and waveguide mode. Light trapped in the substrate (substrate mode) can be readily extracted with microlens arrays on the substrate back [6,7]. In contrast, outcoupling of the waveguide mode is much more challenging compared to that of the substrate mode since an internal light outcoupling structure (ILOS) needs to be built up in the organic-based device. Indeed, most techniques for ILOS, such as light scattering structures [8–11], low index grids [12], textured electrodes/substrates [13], corrugated substrates [14], photonic crystals [15], and microcavity structures [16] are introduced between substrates and transparent electrodes.

However, the rough surface of ILOS can induce an electrical short of devices and a serious damage of soft organic layers. Additionally, the electrical and optical properties of transparent electrodes such as ITO can be deteriorated by introducing ILOS because crystal growth of transparent conductive oxide is remarkably affected by the surface conditions of substrates [17–19]. Moreover, some of these techniques are limited to practical applications due to the complicated fabrication process, strong angular dependency, or poor scalability. To extract both substrate and waveguide modes for maximizing the efficiency of OLEDs, the development of the high performance ILOS which does not cause any damage to organic layers and transparent electrode is of great necessity.

Here, we report the highly enhanced light outcoupling efficiency in OLEDs based on nanoparicle-embedded hole transport layers (NE-HTLs) fabricated by a simple solution process. The NE-HTLs show smoothly formed corrugated surfaces which reduce the total internal reflection at the interfaces of organic layers and transparent electrodes in devices. It is noted that we can avoid damage of organic layers and deterioration of the electrical and optical properties of transparent electrodes since

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[*] Corresponding author.
E-mail address: yhkim113@pknu.ac.kr (Y.H. Kim).

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the ILOSs are prepared on top of the transparent electrode. The OLEDs with NE-HTLs show enhanced power efficiencies by a factor of 1.44 for polystyrene (PS) nanoparticles and 1.78 for SiO_2 nanoparticles, respectively. Additionally, the OLEDs with NE-HTLs show the angle-independent light outcoupling enhancement.

2. Experimental

Fabrication of NE-HTLs: The cross-linked PS nanoparticles (size: ~100 nm) were synthesized by an emulsion polymerization as described in elsewhere [20]. The SiO_2 nanoparticles (size: ~100 nm) were dispersed in water (30 wt%), purchased from S-CHEMTECH. Each nanoparticle dispersion was re-dispersed into poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS) aqueous solutions (AI4083, Heraeus) with a weight ratio of 0.1:1 and 0.3:1 for PS-based NE-HTLs, and 0.05:1 and 0.1:1 for SiO_2-based NE-HTLs, respectively. The nanoparticle dispersions were spin-cast atop indium tin oxide (ITO) substrates at 5000 rpm for 60 s. Subsequently, the spin-coated films were baked at 120 °C for 15 min. The surface morphology of NE-HTLs was examined by scanning electron microscope (SEM, S-2400 Hitachi) and atomic force microscopy (AFM, Icon-PT-PLUS, BRUKER).

Fabrication of OLEDs: Organic layers for NE-HTLs with PS and NE-HTLs with SiO_2 were spin-coated in nitrogen-filled glove box and in air, respectively. NE-HTLs were spun on ITO deposited glass substrate. Poly (9-vinylcarbazole) (PVK), 4,4′-N,N′-dicarbazole-biphenyl (CBP), and the green phosphorescent dopant (Tris[2-(p-tolyl)pyridine]iridium(III) ($Ir(mppy)_3$) were used as the emissive layer (EML). The mixture of PVK:CBP:$Ir(mppy)_3$ (0.45:0.45:0.1 by weight) was dissolved in chlorobenzene at a concentration of 20 mg/ml. The EML solutions were spun onto the NE-HTLs at a spin speed of 800 rpm and annealed at 100 °C for 30 min. 2,2′,2′-(1,3,5-benzenetriyl)tris-[1-phenyl-1H-benzimidazole] (TPBi) dissolved in methanol (0.5 wt%) as the electron transport layer was spun on the EML at a spin speed of 8000 rpm and annealed at 120 °C for 10 min. The thicknesses of PEDOT:PSS, EML, and ETL were 35, 61, and 20 nm, respectively. Finally, LiF (1 nm) and Al (100 nm) were deposited on TPBi via thermal evaporation under high-vacuum conditions. The active area of devices was 6 mm^2. The performances of OLEDs were measured by a goniometer equipped spectroradiometer (CS-2000, Minolta) and source-measure unit system (Keithley Instruments).

3. Results and discussion

To fabricate the corrugated surface of HTLs, we introduce nanoparticles of PS or SiO_2 into PEDOT:PSS thin films. The nanoparticles are dispersed in PEDOT:PSS aqueous solutions and the PEDOT:PSS solutions are simply spin-coated onto ITO substrates. Fig. 1 shows the scanning electron microscope (SEM) images for NE-HTLs embedded with 100 nm-sized PS or SiO_2 nanoparticles, which are denoted as NE-HTL-P and NE-HTL-S, respectively. In contrast to the typical flat surface of PEDOT:PSS thin film without nanoparticles, all NE-HTLs clearly exhibit randomly distributed nanoparticles in the films without remarkable agglomeration. It is confirmed that the PEDOT:PSS films can possess corrugated surfaces by introducing 100 nm-sized nanoparticles. It is observed that the number of nanoparticles increases with increasing the concentration of nanoparticles. NE-HTL-P exhibits more uniformly distributed nanoparticles compared to NE-HTL-S. The small aggregation of SiO_2 nanoparticles in NE-HTL-S is in acceptable range, which does not induce a significant electrical deterioration of devices. Moreover, NE-HTLs have a random pattern across the corrugated surfaces, enabling for efficient angle-independent light outcoupling.

The atomic force microscope (AFM) topography images of NE-HTL-P and NE-HTL-S are shown in Fig. 2. It is observed that the corrugated structures of NE-HTLs are well developed and the morphology characteristics of films are consistent with SEM observations. The root-mean-square (RMS) roughness values for NE-HTL films with 0.1 wt% PS, 0.3 wt% PS, 0.05 wt% SiO_2, and 0.1 wt% SiO_2 are 7.4, 11.4, 7.4, and 9.6 nm, respectively. In contrast, the PEDOT:PSS thin film without nanoparticles show very smooth surface with a RMS roughness of 0.8 nm. It is expected that the higher roughness of NE-HTLs is still smooth not to cause electrical short in devices as reported by several studies [9,21–23]. The smoothly formed corrugated surfaces are expected to effectively change the light pathway, resulting in reduction of the total internal reflection at the interfaces of emissive layers (EMLs). A small aggregation of SiO_2 nanoparticles is also observed in NE-HTL-S as shown in SEM images. It should be noted that the corrugated surfaces are ideally formed without significant spikes and a large aggregation of nanoparticles.

To investigate the light outcoupling effect of corrugated NE-HTLs for OLEDs, we fabricate all solution-processed green OLEDs. HTLs (NE-HTLs), EMLs, and electron transport layers (ETLs) are prepared by spin coating process. The solution processing including ETLs can contribute to mass production of low-cost printed OLEDs. Fig. 3a and b shows typical current density-voltage-luminance (J-V-L) curves for OLEDs with pristine PEDOT:PSS films, NE-HTL-P, and NE-HTL-S. For OLEDs having NE-HTLs, the current densities show a decreasing tendency with increasing the concentration of nanoparticles. It is expected that the insulating nanoparticles incorporated in PEDOT:PSS films hinder the injection of charges from ITO electrodes to emissive layers. Although the reduced hole injection of NE-HTLs, the luminance levels of devices with NE-HTLs do not significantly reduce compared to reference OLEDs without nanoparticles. The enhanced light outcoupling performance by NE-HTLs can be observed in the current efficiency (CE) and the power efficiency (PE) of devices as shown in Fig. 3c–f. For OLEDs with NE-HTL-P, a maximum CE and PE of 34.0 cd/A and 13.8 lm/W are observed at the 0.1 wt% of PS, enhanced by a factor of 1.26 and 1.44 compared to the reference device (CE: 26.9 cd/A, PE: 9.6 lm/W), respectively.

The effect of light outcoupling enhancement is also observed for devices with NE-HTL-S. Both NE-HTL-S films with 0.05 and 0.1 wt% SiO_2 result in the improved CE and PE compared to the reference device (CE: 16.9 cd/A, PE: 5.9 lm/W). The devices for NE-HTL-S fabricated in air result in the somewhat lower efficiency of the reference device compared to that of the reference device for NE-HTL-P, which is processed in a nitrogen-filled glove box. The device with 0.05 wt% SiO_2 shows a highest CE of 29.6 cd/A and PE of 10.5 lm/W, an improvement by a factor of 1.75 and 1.78, respectively. Despite of the inhibited hole injection, the efficiencies of devices with NE-HTL-P (0.1 wt% PS) and NE-HTL-S (0.05 wt% SiO_2) are significantly improved. This enhancement is mainly caused by the fact that the NE-HTLs effectively suppress the total internal reflection at the interfaces between HTLs and emissive layers by corrugated structures. The total and diffuse transmittances of all NE-HTLs as a function of the concentration of nanoparticles are almost identical (not shown). In addition, the refractive indices and absorbance values for pristine PEDOT:PSS films and NE-HTLs do not show a significant difference. These results suggest that the light scattering effect of NE-HTLs is not appreciable reason in the outcoupling enhancement of devices. Fig. 3g and h shows the normalized electroluminescence spectra of OLEDs with NE-HTL-P and NE-HTL-S, respectively. Devices with NE-HTLs exhibit an angle-independent enhancement in luminance attributed to the randomly formed nano corrugated structures on the surface of NE-HTLs. The almost ideal Lambertian emission pattern for Devices with NE-HTLs can contribute to the development of the high performance ILOS with the superior angular emission stability.

4. Conclusions

In conclusion, the high performance internal light outcoupling structures for OLEDs based on nanoparticles-embedded corrugated HTLs are developed by a simple solution process. For creation of surface corrugated patterns of HTLs, various nanoparticles are dispersed in

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