G-1-02 Extended Abstracts of the 2020 International Conference on Solid State Devices and Materials, VIRTUAL conference, 2020, pp413-414

Femtosecond Laser Patterning of Organic Light-Emitting Diode Substrates for the Enhancement of Outcoupling Efficiency

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Abstract

The use of a simple femtosecond laser to fabricate high-precision inverted hole structures at the air/glass interface of OLED substrates, is shown to eliminate the need for lengthy multi-step and complicated outcoupling structure fabrication techniques. Where the inverted hole dimensions are first optimized in via simulation, up to 60.81% enhancement in OLED outcoupling efficiency was achieved as a result of extracted substrate guided modes.

1. Introduction

Since the advent of the first organic-light emitting diode (OLED) by Tang and Vanslyke [1], these devices have made considerable improvements as display and lighting sources. The development of highly efficient emissive materials (for example, thermally activated delayed fluorescent (TADF)) has seen these devices realize up to 100% in their internal quantum efficiencies (η_{IQE}). However, realizing high external quantum efficiencies (η_{ext}) is still limited by the device's low outcoupling efficiency (η_{out}) of ~20% [2], described by eq. (1).

$$\eta_{ext} = \eta_{IQE}\eta_{out} \qquad (1)$$

To enhance the η_{out}, both μm- and nm-scale protruding structures have been implemented in past research efforts at the air/glass and glass/organic interface of the device to suppress the loss of light to substrate and organic waveguided modes, respectively [3]. The fabrication of these structures, however, may include arduous multi-step and complicated fabrication techniques that may be very time consuming. For example, in previous work, the fabrication of a corrugated sapphire substrate to enhance the η_{out} by ~25% included deposition of silica particles via LB-technique, inductively coupled plasma reactive ion itching, followed by silica removal via buffered oxide etching before device fabrication [4].

Advances in laser and chirped pulse amplification (CPA) technology has recently given birth to the femtosecond lasers, which produce high-intensity and ultra-short laser pulses. They have shown established applicability in the medical and industrial fields as corrective ophthalmological surgery (laser-assisted in situ keratomileusis-LASIK) [5] and for high 3D laser lithography, respectively, due to their high precision and minimal damage (smaller heat affected zone compared to lasers with longer pulses). By using a simple femtosecond laser to inscribe inverted patterns the air/glass interface of the OLED, we demonstrate that the need for complicated and time-consuming fabrication methods for outcoupling structures can be eliminated. In this work, we demonstrate that high precision inverted patterned holes created by the femtosecond laser can re-direct substrate guided modes into the forward viewing direction. This leads to an enhancement in the outcoupling efficiency ($\Delta\eta_{out}$) of up to 31.00% and 60.81% in OLEDs without and with PEDOT:PSS, respectively.

2. Experimental Methods

The dimensions of the patterns to be made at the air/glass interface of the OLED were first optimized for depth, diameter and shape on the micrometre-scale via simulation in the program, Setfos 4.6.9 (semiconductor thin film optics simulation software, by FLUXiM), which would produce the best enhancement in outcoupling efficiency.

All substrates were purchased with pre-patterned indium tin oxide (ITO) at the glass/organic interface, where they were used to fabricate four devices, A-D as seen in Fig 1(a). These combinations allow us to analyse the influence of patterned substrates on the η_{out} efficiency among different OLED structures. An Ytterbium doped femtosecond laser (FCPA μJewel DX-0540, IMRA America Inc.) was then used to create the optimized inverted hole patterns (based on the dimensions of the simulation) at the air/glass interface of substrates that would be used to fabricate devices B and D. The laser used a centre wavelength of 1045nm, pulse duration of 450fs and maximum average power of 5W. Afterward, an 85nm-thick PEDOT:PSS layer was then spin coated over the ITO-side of substrates used to fabricate devices C and D, only. Finally, over the ITO- and PEDOT:PSS- side of all substrates, the same organic and metal thin films used to complete the OLED architecture were deposited in an ultra-high vacuum deposition chamber. The layers deposited include α-NPD 90nm/Alq_3 70nm/LiF 1nm/Al 100nm. In summary, device A served as the reference for patterned device B (both without PEDOT:PSS) and device C served as the reference for patterned device D (both with PEDOT:PSS).

3. Results and Discussion

From the simulation, the inverted patterned holes were optimized for maximum η_{out} enhancement by first generating a contour plot out of 256 possible combinations between

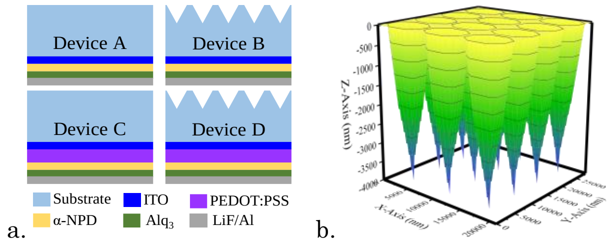

Fig. 1 (a) Structure of OLED devices A, B, C and D used in this study (b) Simulated 3D view of optimized air/glass patterns.

Table I Device Performance in OLEDs Without PEDOT:PSS

Device	J (mA/cm^2)	Voltage (V)	η_{ext} (%)	$\Delta\eta_{out}$ (%)
A	50.51	7.45	1.00	
B	50.78	7.45	1.31	31.00

Table II Device Performance in OLEDs With PEDOT:PSS

Device	J (mA/cm^2)	Voltage (V)	η_{ext} (%)	$\Delta\eta_{out}$ (%)
C	50.86	7.40	0.74	
D	50.22	7.40	1.19	60.81

depth and diameter (ranging from 1 to 15µm). Thereafter, the hole shape was optimized, and the final 3D representation of the most optimized inverted pattern structure, positioned at the air/glass interface of the OLED device is described in Fig. 1. Where the depth, diameter, pitch and shape of the holes arranged in a square lattice are 4µm, 6µm, 7µm and conical, respectively, the simulation predicted a 30.36% η_{out} enhancement ($\Delta\eta_{out}$) in devices without PEDOT:PSS.

Both devices A and B demonstrated similar charge injection characteristics (due to good overlap of current-density-voltage characteristics) and no shift in emission zone (due to good overlap in their forward viewing EL spectra). Thus, any changes in their η_{ext} is a direct result of changes in their η_{out}. From Table I, the patterned device B shows an $\Delta\eta_{out}$ of 31.00% compared to reference device A as a result of the air/glass patterns extracting light originally guided within the substrate into free space (also demonstrating good agreement with the simulation). In planar substrate devices, light is only able to escape into air from the substrate, once the rays incident on the boundary make contact with the glass surface at an angle smaller than the critical angle (θ_c). This phenomenon is described by Snell's law in eq. (2) and is a result of the differences in the refractive index of the two mediums, air (n_2) and glass substrate (n_1) [6].

$$\theta_c = \sin^{-1}\left({}^{n_2}/_{n_1}\right) \qquad (2)$$

Thus, where light was totally internally reflected at the glass/air boundary in device A and thus substrate guided, they now have an opportunity to be outcoupled into free space once the rays meet an angular surface, as is the case in patterned device B. The slant surface of the glass patterns reduces the contact angle made between the rays and substrate, thus allowing the extraction of light at an angle closer to that of the new surface normal.

In reference device C, the absolute value of η_{ext} is expectedly lower than that of device A, as a direct result of lower transparency, due to the thick PEDOT:PSS. This device is thus likely to have a higher percentage of guided modes. Similarly, the η_{ext} between C and D are thus comparable and representative of changes in η_{out} due to good overlap in their current-density-voltage and EL spectra. From Table II, the $\Delta\eta_{out}$ demonstrated in patterned device D, relative to its reference C, is 60.80%. This represents an almost double enhancement compared to the enhancement seen in patterned devices without PEDOT:PSS. Therefore, not only are some of the substrate guided modes extracted as a result of the patterns, but also ~30% lost modes due to the thick PEDOT:PSS can be also be recovered. This result is therefore especially important for solution processed OLED devices where PEDOT:PSS is popularly utilized as the hole injection polymer layer [7].

4. Conclusions

It was demonstrated that the expansion of the applicability of femtosecond lasers to the sphere of the OLED industry has been made possible. By using a simplified one-step method, this laser can inscribe high precision inverted holes at the air/glass interface of the device. These patterns lead to an $\Delta\eta_{out}$ of our devices, up to 60.81% and thus eliminates the need for complicated multi-step fabrication techniques of outcoupling structures.

This work represents the first step toward continuous unity between the two fields towards further enhancing the efficiency in OLEDs. The femtosecond laser has made it possible for the versatile design of patterns not only at the air/glass side of the substrate but can also be applied to the glass/organic side and by extension to transparent thermoplastic substrates.

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