Improved Viewing Angle and Light Extraction Efficiency of Microcavity OLED with Corrugated Capping Layer

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Keywords: OLED, Micro-Cavity, Light Extraction, Corrugated, Thermal Expansion, Viewing Angle.

Abstract: Simple and low-cost materials and process for the formation of light extraction layer of organic light emitting diodes (OLED) are important. However, in case of microcavity-driven OLED, microscale structure for improved light extraction of devices is not easily applicable due to the sever interference with efficiency and viewing angle limitation. In this work, various type of periodic and non-periodic corrugated pattern array for OLEDs were formed by soft lithographic process or spontaneous self-assembly after thermal annealing. Improvement of top emission OLED with microcavity-driven high efficiency were observed, while the angular dependence of light emitting spectra (viewing angle characteristics) was reduced at devices with larger features of corrugated non-periodic capping layer structure. Optical properties of devices were investigated in terms of the scale of periodic patterns and optical cavity effect. Methods in this work could be utilized as an effective tool for managing microcavity-driven performance of high efficiency OLEDs.

1 INTRODUCTION

There exist various technologies for a light extraction (LE) scheme for organic light emitting diodes (OLEDs). However, for the successful commercial application for high-performance OLED display with micro-cavity structure, the restriction on the cost of manufacturing, directionality of color or viewing angle, and the requirement to maintain a high-quality image are severe. For a top-emitting OLED (TOLED), which is a useful platform of high-aperture OLED display with high-resolution mobile and non-transparent flexible substrates, color and spectra are significantly sensitive to the intrinsic micro-cavity effects. Recovering of the optical loss from surface plasmon (SP) is also effective strategy for an enhancement of light extraction efficiency, and a method of grating coupling by a generation of corrugate the organic/metal interface were found to be effective. Several kinds of periodically corrugated patterns were proposed, showing a reduction of light loss by a scattering of SPs (Koo *et al*, 2010, Koo *et al*. 2011, Yin *et al*, 2016).

In this work, we have introduced some of the simple fabrication processes for the microscale LE structure formation. Several patterns of regular and irregular photonic structures were introduced with controllable scale. In case of the generation of a quasi-periodic corrugation patterns with broad size distribution for LE structures of top-emitting OLED, formation of corrugated patterns is spontaneously driven by the mismatch of thermal expansion coefficient of the organic and polymeric layers, which are easily fabricated by simple deposition process. The luminous efficiency of device with corrugated patterns was generally increased, where the change of the spectral characteristics and color stability depends on a status of micro-cavity for specific TOLED devices. Experimental data for different scale of buckling patterns at top emitting OLED were investigated with the wavelength-dependent grating pitch and scattering order and optical simulation, showing that increased scale/pitch of corrugated patterns at capping layer resulted in a suppression of viewing angle variation for microcavity OLEDs.

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Chin, B. D., Yoo, J.-Y. and Jo, S. M.
Improved Viewing Angle and Light Extraction Efficiency of Microcavity OLED with Corrugated Capping Layer.
DOI: 10.5220/0013311700003902

In *Proceedings of the 13th International Conference on Photonics, Optics and Laser Technology (PHOTOPTICS 2025)*, pages 142-146
ISBN: 978-989-758-736-8; ISSN: 2184-4364

2 EXPERIMENTS AND RESULTS

2.1 Corrugation Pattern Formation: Internal and External LE Layers

1D regular photonic structures were generated from master molds, followed by soft lithographic process (Fig 1a), while the irregular 2D corrugated patterns for LE structures capping layer for top-emitting OLED were described in Fig 1b. More detailed information of process such as thermal treatment could be found elsewhere (Koo *et al*. 2021).

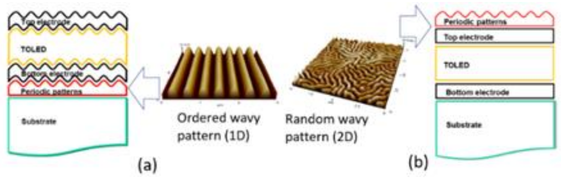

Figure 1: Comparison of the LE structures for TOLEDs in this work (a) Ordered wavy 1D formed on the bottom-substrate of TOLED expecting very weak microcavity (b) Random 2D wavy pattern on the top electrode of TOLED; maintaining the micro-cavity.

2.1.1 1D Regular Pattern

The pattern of 1D wavy type was fabricated through thermal reflow process of positive photoresist (PR). The depth of the pattern was adjusted according to the spin coating condition of the positive PR. Si wafer-based master mold with original pattern (height 800 nm, width 800 nm, pitch 1.6 um) was ultrasonically cleaned using Acetone and IPA and blown with N_2. Then dried in an oven at 180°C for 1 hour. Positive PR (ma-P1205, Microresist corp.) was dropped on the cleaned master mold and maintained for 2 min, then spin-coated at 2000 rpm for 60 seconds. When the spin coating was completed, a wavy pattern was formed by heating treatment for 10 min on a hot plate of 100°C. The polydimethylsiloxane (PDMS) was mixed with the curing agent at a ratio of 10:1 and poured onto the wavy pattern. It was stored at room temperature for 24 hours and removed bubbles inside. The PDMS was thermally cured by storing in an oven at 60°C for 4 hours. The PDMS replica mold was separated from the master mold and ultrasonically cleaned for 10 min in a 1:1 mixture of acetone and isopropyl alcohol (IPA). Then, using N_2, it was dried in an oven at 180°C for 1 hour to produce PDMS replica mold. The pre-cleaned glass with PR-spin-coated at 4000 rpm for 60s was pre-baked on a 100°C hot plate for 1 min. The PDMS replica mold with wavy pattern was transferred to pre-baked PR by thermal nanoimprinting lithography (NIL) process. The thermal NIL process was carried out on a 100ºC hot plate and the pressure was applied for 1 hour using a weight of 1kg or 3kg. Wavy patterns for devices B, C, and D (atomic force microscopy - AFM image as seen in Fig. 2b) were prepared by controlling the rpm of the spin coating process at 2000, 2250, and 2500 rpm.

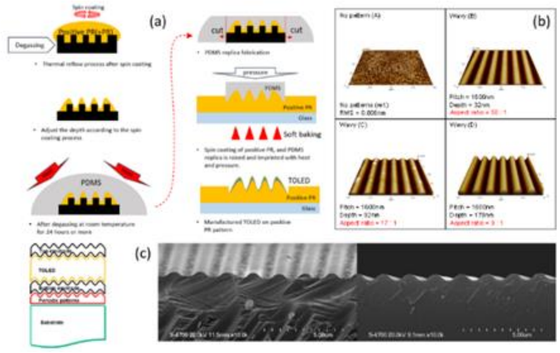

Figure 2: (a) Schematic fabrication process of a wavy patterned TOLED by thermal imprinting (b) scale of the 1D wavy patterns for TOLED device fabrication (c) Cross-sectional SEM image of wavy pattern produced through thermal reflow process (magnification: 10k), (left) using 1 kg, (right) 3 kg weight.

2.1.2 2D Irregular/Corrugated Pattern

At least two organic layers (typically for 35nm/15nm-thick bi-layers) were deposited by thermal evaporation at a high vacuum system with a base pressure below 1 x 10^{-6} torr without breaking the vacuum on top of the TOLED devices with transparent top cathode. After deposition of bi-layer, the sample was thermal treated at 80ºC for 10min in the N_2 chamber for thermal expansion stage. Because the glass transition temperature of other organic materials (charge transporting and emission layers) in TOLEDs were higher than 80ºC, almost no change of device without 2D pattern after this thermal annealing. Followed by the heating, the sample was cooled to ambient temperature by keeping in the N_2 chamber for 5min. The difference in coefficient of thermal expansion (CTE) between the bi-layer on top of transparent electrode of TOLED generated a spontaneous buckling structure.

2.2 Fabrication of Microcavity OLEDs

Microcavity OLED with top-emission structure were fabricated by thermal evaporation onto pre-cleaned glass substrate. All of the following deposition process were performed through a thermal evaporation without breaking vacuum. Aluminum layer was deposited on glass substrate as a reflective anode electrode having a thickness of 100 nm. Afterwards, 2 nm-thick MoO_3 layer for hole injection and buffer, 28 nm-thick TCTA layer for hole transport, 30 nm-thick EML layer of doped co-host system (TCTA : TmPyPB : $Ir(ppy)_3$ = 0.465 : 0.465 : 0.07), 25 nm-thick TmPyPB layer for electron transport, 0.5 nm-thick LiF layer and 1 nm-thick Al layer for electron injection, and 18 nm-thick Ag layer for composite semi-transparent cathode electrode were deposited. Surface morphology of sample was measured by atomic force microscopy (AFM, Veeco Instruments) and scanning electron microscope (SEM), etc. Fabricated TOLEDs were characterized using a source meter (Keithley 2400) with a spectrometer (PR655, Photo Research) and custom-built goniometer for angular dependence measurement of light emission. All electroluminescence spectra and angular emission patterns were recorded at a constant current density of 10mA/cm^2 with a spectrometer

2.3 Results of Device Data: Improved LE and Viewing Angle Behavior

Fig 3 (a) illustrates current density - external quantum efficiency behavior of TOLED with 1D wavy underneath of entire layers. The devices with wavy pattern showed slightly higher current density than references. At the inflection point of these sinusoidal patterns, the thickness of the organic layer is partially thinned, which might result in the formation of a stronger local electric field. Luminance tended to increase as the depth of the wavy pattern increases, however, EQE values are only slightly increased at device B (low-height 1D wavy) compared to reference device A, showing only similar values of 7.73 and 7.56% for devices with higher 1D patterns (device C and D, respectively). Fig 3b shows normalized spectral intensity in the normal direction of the emitting surface. The sinusoidal wavy pattern gradually changes the microcavity length of the TOLED. The thickness of the cavity length (organic layer) became thinner at the inflection point and thickens at the trough peak (Liu *et al*. 2012). With the height of wavy patterns inside TOLED increases, the micro-cavity effect starts to be reduced, so that angular dependence of the spectrum for device D is less-sensitive compared with reference device A. However, overall performance of wave LE layer was not great for these structures.

Fig 4 illustrate the representative image obtained by AFM for 2D random wavy patterns that organic bi-layers with thermal expansivity difference generates. The pitch and depth of patterns could be systematically controlled, depending on the annealing temperature and time. The Si wafer and the buckling structure formed on the top of the TOLED are shown in Fig 4a and 4b, respectively. For example, 35nm N,N'-Bis(3-methylphenyl)-N,N'-diphenylbenzidine (TPD) and 15nm tris(8-hydroxy-quinolinato) aluminum (Alq_3) deposited and annealed at a temperature of 80°C were shown. The both measured patterns had 750 nm pitch and 30 nm depth.

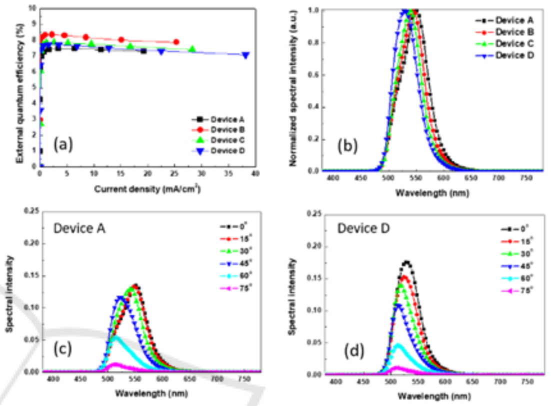

Figure 3: (a) Current density v. EQE, (b) spectral response (c)(d) angular dependence per each viewing angle for device A and D, without and with 1D micron scale wavy pattern. Devices are identical with structures in Fig. 2.

Figure 4: AFM image of 2D buckling structure formed by spontaneous thermal expansion/compression (a) on the Si wafer substrate and (b) on the TOLED device (scan size: 10 x 10 μm).

Since the formation of buckling 2D wavy patterns formed on top of the transparent cathode of TOLED does not affect the thermal state of the underlying organic layer of devices, micro-cavity of those TOLEDS were well maintained. Devices in Fig 5a showed; the layout of LE bucking structures, where device A (EQE 12.4 represents the reference without LE, device B shows non-bucking LE with same organic bi-layer. For devices of C, D, and E, wavy pattern pitch/depth is 750/30, 1000/45, and 1150/64 (nm), respectively. The improvement of external quantum efficiencies were significant in case of TOLEDs with 2D buckling, where the most optimized devices E showed 21% (reference A and w/o buckling LE device B shows EQE of 13.5 and 17%; see Fig 5b). More than 50% of EQE was obtained for device with largest 2D wavy LE structure. Such a result, observed in case of optimum buckling structure formed on top of the translucent electrode of device, can be explained by the suppression of light loss by the SP mode at semi-transparent metal/air interface without losing the control of polarization of light. Moreover, the shift of the maximum peak of electroluminescence for buckling patterned device was less than 4nm with the viewing angle range of 0 to 75 degree (>20nm shift for control device), securing a negligible change of the spectral characteristics and color stability (robust viewing angle behavior as for cavity-based device, comparable results with Kim *et al*. 2017) as seen in Fig 5c.

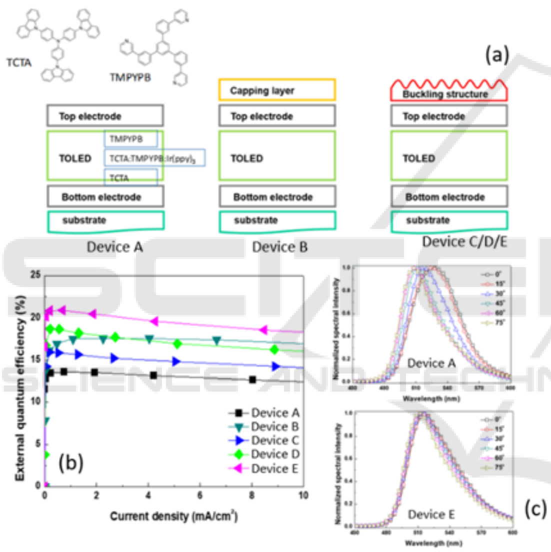

Figure 5: (a) Layout of TOLEDs; reference, flat capping layer, and 2D buckled layer (same bi-layer) (b) current density vs. EQE for each TOLEDs with different flat/buckling LE structures (c) comparison of the angular dependence for reference device A and 2D buckling device E.

3 CONCLUSIONS

As for overall evaluation, corrugated "wavy" patterns at a micron-scale enhances the viewing angle, which is a significant advantage for display technologies. Moreover, luminous efficiency was improved by more than 50% for green phosphorescent TOLEDs as long as they maintained less-sensitive viewing angle dependence and stable color purity. It can be noted that spontaneous formation of 2D wavy buckling structures through thermal deposition and annealing is simple and potentially cost-effective.

For 1D corrugated "wavy" patterns at a general micron-scale, as the aspect ratio of the pattern becomes larger, the interference phenomenon became complicated and the viewing angle was improved. However, efficiency was increased only at limited condition of low aspect ratio 50:1 to 17:1, where the larger patterns break the micro-cavity of TOLEDs. Therefore, this might be a limitation restricting the practical application of this method for the robustness of commercial process.

A simple process for the spontaneous formation of 2D wavy buckling structures generated patterns with pitches of 750 ~ 1100 nm. These randomly directed patterns were formed on top of the cathode (semi-transparent) of TOLEDs by the thermal deposition and annealing process, which did not affect the thermal and transition status of the other organic layers.

Even though the interference phenomenon is rather complicated, and larger patterns breaking the micro-cavity of TOLEDs could lead to structural integrity issues and affect the overall device performance, the improvement of the luminous efficiency more than 50 % of the reference shown at green phosphorescent TOLED will have a strong impact on the progress of device optimization. Since this method ensures less-sensitive viewing angle dependence and stable color purity as well as efficiency boost-up, this approach can be applied various kinds of devices having strong micro-cavity structures, such as TOLEDs with stable color purity and OLED lasers.

ACKNOWLEDGEMENTS

This study was supported by the Industrial Strategic Technology Development Program funded by the Ministry of Trade, Industry and Energy (MOTIE), South Korea (RS-2024-00417913) and Basic Science Research Program Through National Research Foundation of Korea (NRF) funded by the Ministry of Education, Science and Technology (2022R1F1A1074725)

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