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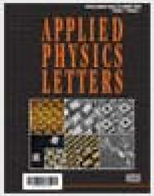

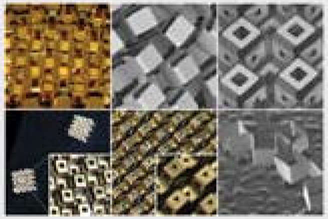

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Applied Physics Letters

Micro-pixel array of organic light-emitting diodes applying imprinting technique with a polymer replica

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Tae Hyun Park,[1] Young Min Kim,[1] Young Wook Park,[1] Jin Hwan Choi,[1] Jin-Wook Jeong,[1] Ki Young Dong,[1] Kyung Cheol Choi,[2] and Byeong-Kwon Ju[1]

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Efficient micro-pixel array of small molecule organic light-emitting diodes (OLEDs) has been fabricated by an imprinting technique which uses a polymer replica. To confirm the effect of the oxygen plasma for removing the residual layer, the performance of two kinds of OLEDs with varying thicknesses of resin as the micro-pixel array, have been compared. The measured results of the OLEDs have shown comparable device performances that are significantly characterized depending on the residues on the substrate. The performance of enhanced device has achieved efficiencies of 3.6 cd/A and 1.9 lm/W at 20 mA/cm^2. ©2009 *American Institute of Physics*

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Organic light-emitting diodes (OLEDs) have been attractive attention for the next generation of display applications, especially in small display devices, such as mobile electronics and flexible displays, because of their fascinating properties of low power consumptions, high contrast ratios, and flexibility.[1,2] Also, it is expected that OLEDs with micro-pixels could be used in high-resolution displays, for describing clear pictures, such as head mounted displays or a variety of medical displays. To define active areas, conventional OLEDs are fabricated by photolithography but this includes a wet process that is unsuitable for flexible substrates as they can be easily damaged by it.[3] Alternatively, imprinting has several advantages compared to conventional photolithography, such as low cost, high resolution, high throughput, and easy processing.[4–8] Also, the imprinting process does not need a wet process or high-energy radiation and has no optical diffraction limit.[9] Nevertheless, imprinting lithography is still limited in its ability to control the residual layer, and this is proving to be an obstacle to the next process. To complement the residual layer problem, a variety of processes have been developed to minimize the residual layer near-to-zero or to do etchless process.[10,11]

In this paper, patterning of micro-pixel array has been experimented, with changing thickness of the residual layer, and different resins for removing the residue completely with oxygen (O_2) plasma from the substrates.[12] Finally, the micro-pixel arrays, fabricated with a thin resin and a thick resin, have been compared to define the characteristics of the OLEDs depending on the residue. All the processes are finished in a dry process, with low temperature. This study demonstrates a basic technique for plastic substrates, which often cause severe damage in the wet process.

Moreover, an alternative stamp, polymer replica is used, which could be used repeatedly to give a low cost process.[13] In order to fabricate the stamps, a mold has been used, which is composed by poly(dimethylsiloxane) (PDMS), commercially available from the Dow Corning Co. This is duplicated from the positively micro-patterned silicon dioxide master stamp, which is fabricated by conventional lithography on the silicon substrate. The polymer stamp is composed by an ultraviolet (UV) sensitive hybrid polymer material, namely Ormocomp, commercially available from the Micro Resist Technology Co. As shown in Fig. 1, the polymer stamp is fabricated in regular sequence. The pitch and diameter of the pattern are 12 and 8 μm, respectively [see Fig. 2(a)] its height is 400 nm. The polymer stamp is then treated with a self-assembled monolayer (SAM) to reduce adhesion between the stamp and resin. Octadecyltrichlorosilane (OTS) has been used as the SAM, which changes the hydrophilic properties of the surface to hydrophobic properties of surface on the stamp, by immersion in an OTS containing solution.[14]

Two resins have been applied as the micro-pixel arrays, to define the effect of the imprinting, depending on the thickness of the layer. First, a resin is found (MR I-7030, Microresist Technology Co.) characterizing low glass transition temperature (T_g=60 °C), for applying the low temperature process. Next, AZ series of photosensitive polymer resin (AZ 1512) is chosen as a thick layer. Then, each resin is spin coated on an indium tin oxide (ITO) coated glass substrate, and baked on a hotplate. The thickness of the MR I-7030 is 300 nm. This is thinner than the actual height of the pattern whereas the thickness of the AZ 1512 is 1.4 μm thicker than the height of the pattern. After prebaking of the substrate, it was set on a stage in the imprinter equipment. The stamp has been applied to the substrate, and then heated over the glass transition temperature (T_g) of the resin. After embossing the stamp and the substrate for 10 min at 140 °C, the imprinted micro-pixel array of the layer was demolded below $T \sim 60$ °C. The designed micro-pixel array of OLED devices with a large area 1×1 cm^2, and patterned by thermal imprinting on the ITO glass substrates. In order to expose the ITO surface for the anode connection, the thin residual resin on the imprinted layer is removed by O_2 reactive ion etching (RIE) [Fig. 2(b) and 2(e)]. For the fabrication of the OLEDs, a hole transporting layer, n,n'-bis(naphtha-1-yl)-n,n'-bis(phenyl)-benzidine, emitting layer, tris(8-hydroxyquinolinato)aluminum, and cathode layer, lithium fluoride

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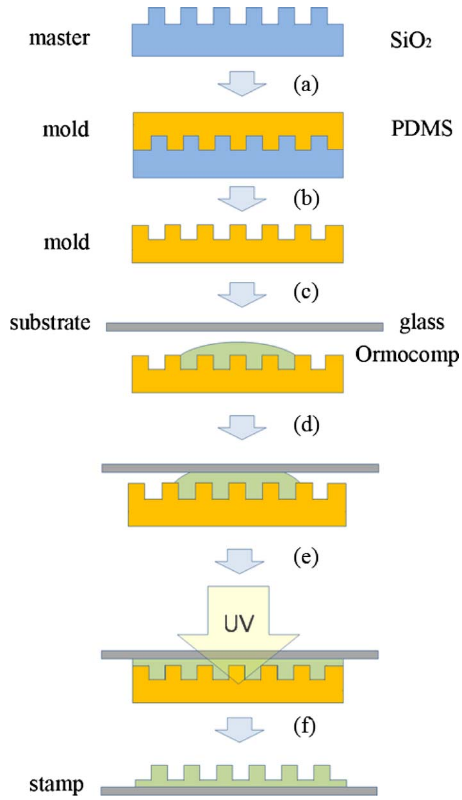

FIG. 1. (Color online) Schematic illustration of the fabrication of a PDMS mold, and a hard stamp by UV imprinting. (a) The PDMS is poured on the master, and then cured in an oven to reverse the patterns of the master. (b) The PDMS mold can then be easily peeled off from the master since the PDMS has the property of elasticity and low surface energy. (c) After the fabrication of the mold, the UV curable polymer is dropped onto the PDMS mold. (d) To form micropatterns into the UV curable polymer, Ormocomp is closed with a glass as a substrate of the stamp. (e) UV light exposing and curing is performed with applying force by a UV imprinter. (f) Finally, a polymer stamp is fabricated, with the dimensions 1×1 cm^2.

and aluminum, are deposited on the substrate consecutively, by thermal evaporation. The fabricated devices are characterized using PR 670 and Keithley 237 for the current versus voltage characteristics and efficiency.

Figure 3(a) shows an image of non-uniform emitting OLED pixels with AZ 1512 (device-A), driven at 18 V, observed under an optical microscope and pictured by a CCD camera. Although the imprinted pattern has been well formed, some parts of the electrodes are covered with residues of granules. Even though sufficient etching time was performed, there remained residues since the resin is polymerized within an O_2 plasma [see Fig. 2(c)].[15] Since the granules were highly etch-resistant, the width of the micropixels was expanded through the over-etching time, and was bigger than the width of the stamp pattern. The thick resin improves to form a pattern as the role of buffer layer. However, residues are an issue in degrading the performance of the device such as luminance efficiency, power efficiency, and life time. On the other hand, Fig. 3(b) shows an image of 8 μm pixel arrays with MR I-7030 (device-B) at 5 V. Since the micro-pixel array has been imprinted with the hard polymer replica, there is no distortion of the patterns. Also, the residual layer is minimized to less than 80 nm, to complement the fact that the resin is polymerized with O_2 RIE.[15] Eventually, the micro-pixel array is demonstrated without residues [see Fig. 2(f)], so that the OLED is fabricated uniformly and emitting regularly.

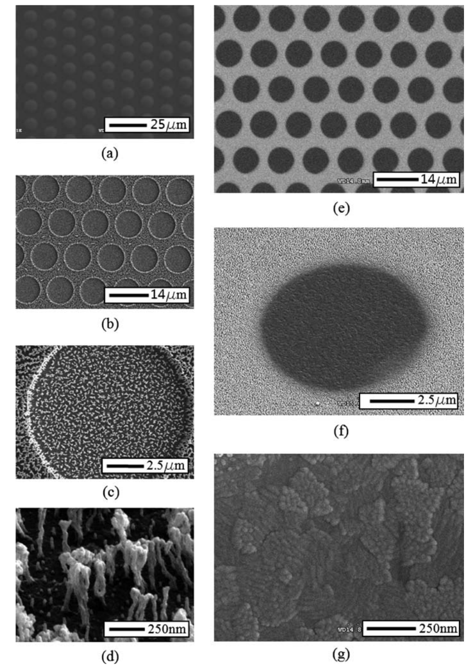

FIG. 2. SEM images of (a) a fabricated stamp (b) imprinted 8 μm patterns on AZ 1512 after exposing of electrode by O_2 RIE. (c) A pixel array made from AZ 1512. (d) An electrode image (AZ 1512) after O_2 RIE. (e) Imprinted 8 μm patterns on MR I-7030 after exposing of electrode by O_2 RIE. (f) A pixel array made from MR I-7030. (e) An electrode image (MR I-7030) after O_2 RIE.

Figure 4(a) shows characteristics of current-voltage of an OLED device patterned micro-pixel array, which has been fitted by the area driven by the injected current. The turn-on voltage of the device B is 3 V, and the current density is 3.2 mA/cm^2. The turn-on voltage is lower than that for conventional OLED devices because of the effect of the O_2 plasma during the etching of the residual layer on the ITO electrodes.[16] On the other hand, the device A shows very low characteristics. The turn-on voltage is 6 V, and the current density is 0.1 mA/cm^2. Because of residues on the electrode, the nanoparticles caused a decrease in the charge injection ability. The ITO/organic interface affect the properties of carrier injection and efficiencies to the device. After applying a high electric field to the device A, lots of dark spots increased, and the electrical properties degraded through nanoparticle residues of the interface between the electrode and the layer. The protrusions and hollows of residue induced a high electric field on the ITO/organic interface, therefore, the turn-on voltage increased.

Figure 4(b) shows the luminance efficiency of OLEDs both with and without residual layers, respectively. The device B characterizes the higher luminance efficiency (3.65 cd/A) than that of the device A (1.46 cd/A) depending on the

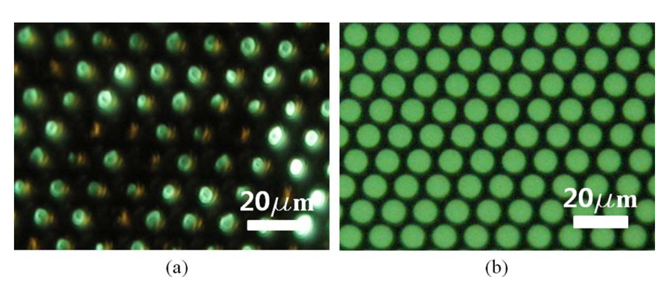

FIG. 3. (Color online) Optical microscopy images of emitting OLED with micro-pixel array using (a) AZ 1512 and (b) MR I-7030.

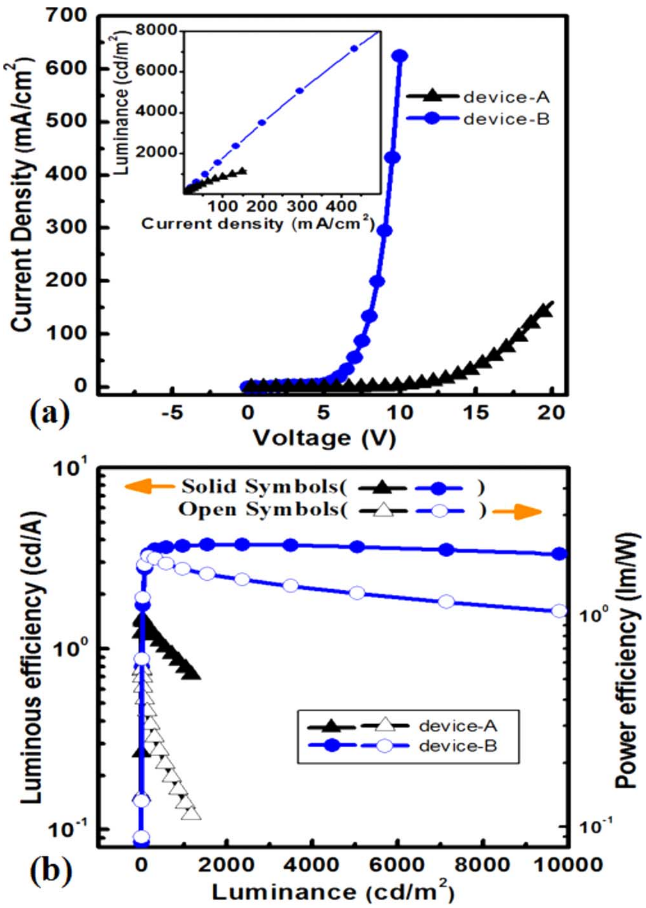

FIG. 4. (Color online) (a) Current density-voltage characteristics, luminance and (b) luminous efficiency and power efficiency.

residues on the electrodes. Figure 2(g) shows the scanning electron microscope (SEM) image of the ITO electrode of device B without residues. However Fig. 2(d) shows the SEM image of many protrusions and hollows (device A), which would be obstacles of characteristics to the devices. Since the local electric field, concentrated on the ITO/organic interface, increased the leakage current, the power efficiency is very poor, and dramatically decreased from 0.61 to 0.11 lm/W increasing the current density. Therefore, removing the residual layer completely is very important for the characteristics of the OLED devices.

In addition, a polymer replica was applied, which combined the benefit of the PDMS and hard stamp. First, PDMS stamps are suitable for a repeatable process since they are characterized by their low adhesion and their elasticity. However, controlling the submicron pattern with PDMS stamps is limited due to expansion of the stamps with high pressures and temperatures. The original patterns might be distorted and unclear on the boundary of the patterns. Contrasts with elastic stamps, hard stamps are not limited by distortion of the pattern, but they are characterized as brittle and expensive. An alternative stamp was made, in order to make patterns with a repeatable and cheap process. As the PDMS was applied to a mold, stamps could be made easily without a residual layer between the mold and the stamp. In addition, the hard stamp made from Ormocomp was able to pattern without any distortion. Also, the surface of the hard stamp was treated as hydrophobic by SAM, reducing adhesion between the stamp and the resin enabling repeatable use of the imprinting process.

The OLED of 8 μm size pixel has been demonstrated using thermal imprinting with a polymer replica. The performance of two kinds of OLEDs was compared, having a micro-pixel array with thinner and thicker resin than the height of the stamp pattern. The measured results of the OLEDs have shown significantly different characteristics, depending on the residues on the electrode. Since the poor conductivity and current leakage of the residual layer degrades the characteristics of a device, residues should be removed for enhanced OLED performance. These limits have been improved, to use thin thermal resin, and to minimize the residual layer to less than 80 nm.

All of the experimental processes have been performed in a wet-free process and a low temperature process. Generally, plastic substrates are composed of polymers, which are easily damaged by the organic solvents as well as the wet processes, and are characterized with a low T_g, limited by a high temperature process. The method experimented here could apply plastic substrates for OLEDs, and would be a basic process for producing flexible displays. Therefore, this imprinting technique is proposed for application to the formation of patterning, based on low temperature, wet-free, repeatable, and low cost processes for the manufacture of high-resolution future flexible displays.

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