High-resolution organic polymer light-emitting pixels fabricated by imprinting technique

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We have developed an approach to fabricate pixelated organic polymer light-emitting devices (OPLED) using an imprinting technique. The pixel array pattern was first defined in an insulating polymer layer on indium tin oxide glass by direct imprinting, followed by the spin-coating of OPLED polymers and cathode metal deposition. We demonstrated successful fabrication and operation of OPLED pixels of sizes from 50 μm down to 2 μm. Optoelectronic characterization is performed on these devices, and measured results show comparable device performance with OPLED pixels patterned by other methods. This fabrication scheme holds many merits such as easy to process, low-cost, high yield, expandable to flexible substrate, capable of repeated imprinting for large area arrays, and the potential to pattern submicron and nanoscale organic polymer light emitters. [DOI: 10.1116/1.1515307]

I. INTRODUCTION

Organic light emitting devices (OLEDs) have attracted enormous research interests and activities during the last decade and will continue to do so in the future because of their fascinating properties and promising applications. OLEDs can operate with a low power consumption and are highly luminescent, with emission spectra covering the whole range of visible light. The simple technique associated with the processing of organic polymer based OLED (OPLED), along with the aforementioned properties make OPLEDs especially attractive in many applications. OPLEDs with large pixels can be used in next generation flat-panel displays, while OPLEDs with small pixels may find use in high-resolution microdisplays, and as light sources in microfabricated photonic devices and systems. Various fabrication methods of OPLED pixels have been reported in literature, such as patterning of electrodes by photolithography,[1] direct photoconversion of a precursor to PPV,[2] soft lithography patterning of hole transport layer,[3] hot microcontact printing of self-assembled monolayers,[4] direct patterning of emissive polymer with solvent-assisted micromolding,[5] and ink-jet printing.[6] Nanoimprinting is an emerging nanolithography technique that combines many attractive characteristics such as low cost, high resolution, and high throughput.[7] Previously it has been applied to pattern submicron grating structures directly in an electroluminescent material.[8] In this article, we report the demonstration of electrical-injection OPLEDs fabricated by the imprinting technique.

The imprinting technique uses a hard mold with protruded patterns defined on its surface to imprint into a polymer layer cast on a substrate. The imprinting process is achieved under an applied pressure and at a temperature higher than the glass transition temperature of the polymer film.[7] Numerous applications based on imprinting have been demonstrated in the past. For example, many electronic, optoelectronic, and photonic device structures have been fabricated by the nanoimprint technique. These include a nanoscale field effect transistor,[9] high density magnetic disks,[10] a metal–semiconductor–metal photodetector,[11] a broadband waveguide polarizer,[12] and nonlinear optical polymer nanostructures.[13] Further improvement of nanoimprint technology, such as developing resist materials,[14,15] achieving alignment capability,[16] and approaches aimed at defect reduction during nanoimprinting,[17] will likely push the imprint technology into commercial applications for fabrication of integrated electronic and photonic devices.

II. EXPERIMENT

A. OPLED pixel array fabrication

Our approach of applying imprint technology to the fabrication of pixelated OPLEDs is to first use imprinting to create a template of pixel array patterns in an insulating polymer material (Fig. 1), and then deposit OPLED polymers over the template. Only the pixel regions that provide a conduction path to the organic polymer will light up when driven by an injected current. The pixel patterns on the mold were fabricated on a silicon oxide layer thermally grown on a silicon substrate. The pixel patterns on the mold were first defined by conventional photolithography, and then transferred into a silicon oxide layer by reactive ion etching (RIE) using CHF_3 and O_2 gases. Typical pixel patterns on a fabricated mold that has 10 μm pixel size is shown in Fig. 2(a). The mold is then coated with perfluorodecyltrichlorosilane surfactant to reduce its surface energy, which provides easy mold release property after the imprinting process. To create an insulating pixel template, a thin film of poly(methylmethacrylate) (PMMA, Mw = 15 000) was first spun onto the indium tin oxide (ITO) glass substrate. Then the mold and the PMMA coated ITO glass substrate were brought into

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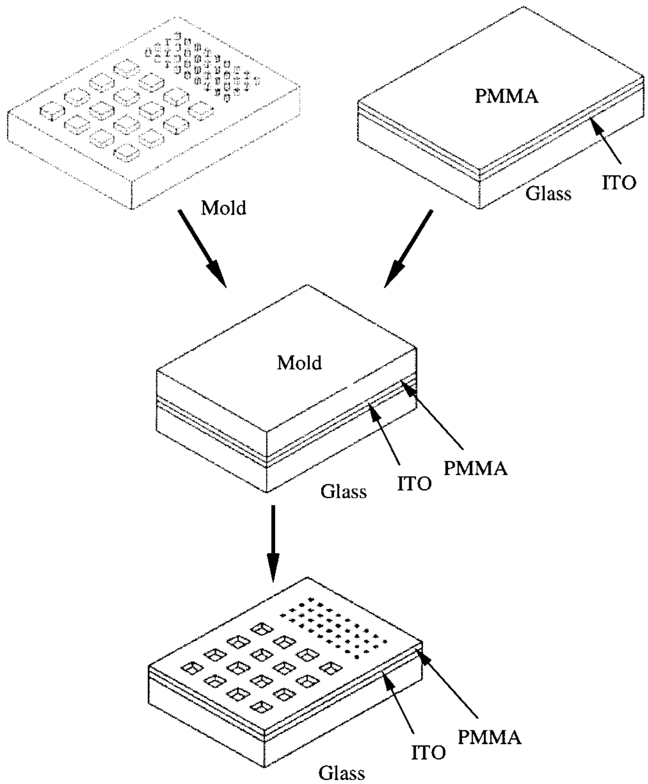

FIG. 1. Schematics of fabricating an OPLED pixel template in PMMA by imprinting.

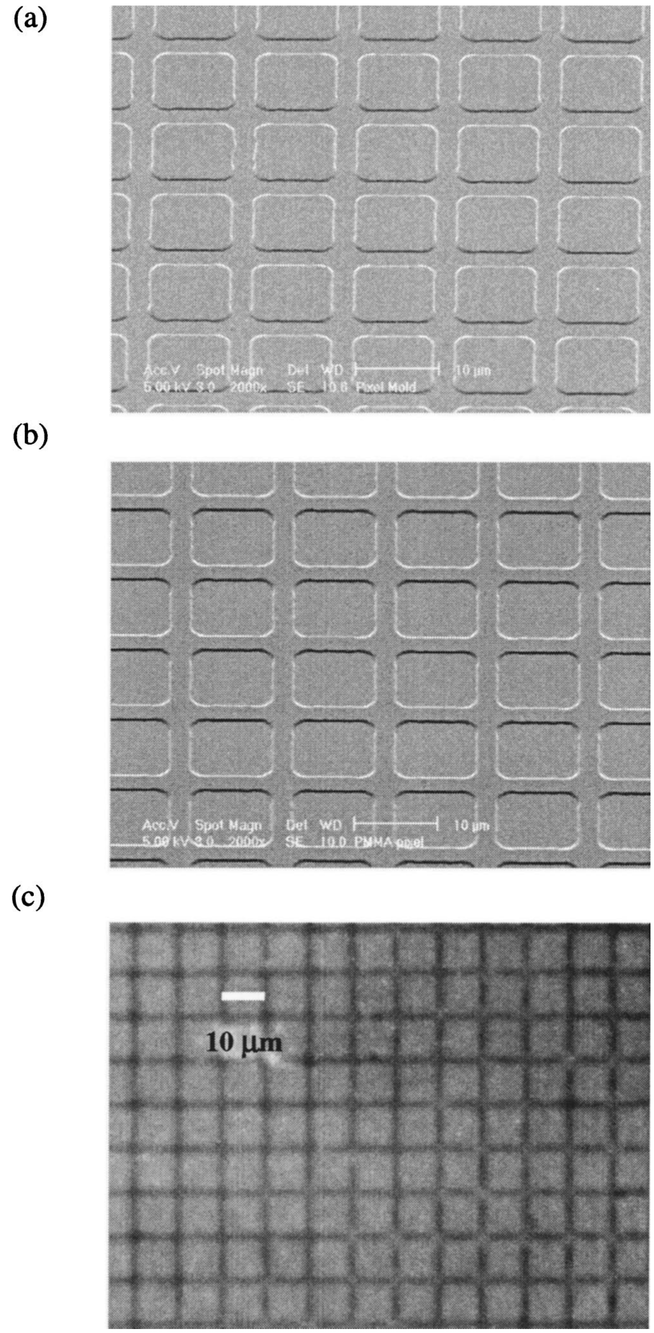

FIG. 2. Scanning electron micrographs of (a) pixel patterns on the mold and (b) imprinted pixel template in PMMA, and (c) optical micrograph of lighted OPLED arrays taken by a charge coupled device camera mounted on a microscope. The pixel size is 10 μm, and the light emission from the pixels is red.

contact and transferred into a custom-made hot press that can apply pressure and heat. The mold and substrate were kept at 50 kg/cm^2 pressure and 175 °C for 5 min. They were cooled to ambient temperature before separation. The pixel pattern on the mold was physically imprinted into the PMMA, and the thin residual PMMA within the imprinted pixels was then removed by O_2 RIE to expose the ITO surface for anode connection [Fig. 2(b)]. Next, a hole transporting layer, poly(3,4-ethylenedioxythiophene)-polystyrene sulfonate (PEDOT-PSS), and a red emissive polyfluorene derivitive polymer layer were consecutively spin-coated and cured. The cathode electrodes were formed by thermal evaporation of aluminum under the high vacuum ($\sim 10^{-7}$ Torr) through a shadow mask with a rectangular area of 2×3 mm^2. The fabricated OPLEDs were then cured in the vacuum oven at 90 °C for 1 h before optoelectronic measurements.

B. Optoelectronic characterization

The luminous flux and current versus voltage characteristics were measured in air with a programmable voltage source (Keithley 230), an electrometer (Keithley 617), and an IL1700 Research Radiometer from International Light. The fabricated devices were mounted on one port of the integrating sphere, which is optically separated from the detector port by specifically designed baffles (Fig. 3). All the electrical and optical measurements were performed at the same time by automatic control through a computer. In our fabricated devices, the Al pad covers pixels having sizes ranging from 2 to 50 μm. So the measured characteristics are the overall properties of the pixel arrays with different sizes. Knowing that such measured properties do not represent the specific property of pixels with a certain size, we only intended, through this study, to get qualitative information about the performance of the OPLED pixels that are fabricated using this technique.

III. RESULTS

Upon applying voltage bias, bright red light emission was observed from clearly defined pixels that have sizes ranging

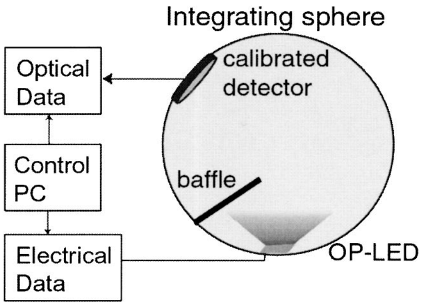

FIG. 3. Schematics of the optoelectronic property characterization system.

from 50 down to a few μm. Figure 2(c) shows the photographs of light-emitting OPLED pixels with 10 μm size that were taken by a charge coupled device camera mounted on an optical microscope. The smallest OPLED pixels in our experiment have a size of 2 μm (Fig. 4), only limited by photolithgraphically defined pixel size on the mold we currently used.

Figure 5 shows the current and luminous flux versus applied voltage characteristics of fabricated OPLEDs in linear and semilogarithmic scales, respectively. The turn-on voltage and the corresponding current are $\sim$5.5 V and $\sim$30 μA, respectively, which are defined at the lower limit of sensitivity (10^{-6} lm) for our measurement setup. The luminous flux up to $\sim 10^{-3}$ lm has been obtained for the OPLEDs where the applied voltage is limited to 10 V to allow repeated measurement without device degradation. However, we expect that the luminous flux of up to 10^{-2} lm, which is equivalent to the luminance of several thousands cd/m^2 for our devices and the measurement setups, can be easily obtained without any device degradation.

Even though nonemitting OPLED pixels also exist in our devices due to pattern defects created during the imprinting process, the overall device yield is around 80%. We are currently working on reducing the defects in the imprinting process. At a high bias level of $\sim$25 V, we observe that the contrast of pixels to the background decreases, especially for the small pixels. 2 μm pixels become indistinguishable from the background after being biased at 25 V for tens of sec-

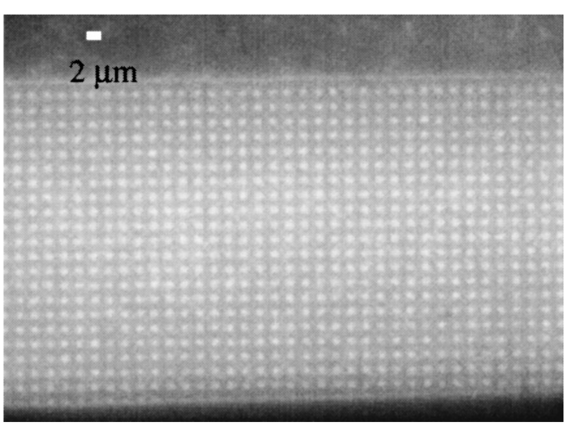

FIG. 4. Optical micrograph of light emission from the smallest size (2 μm) pixel arrays.

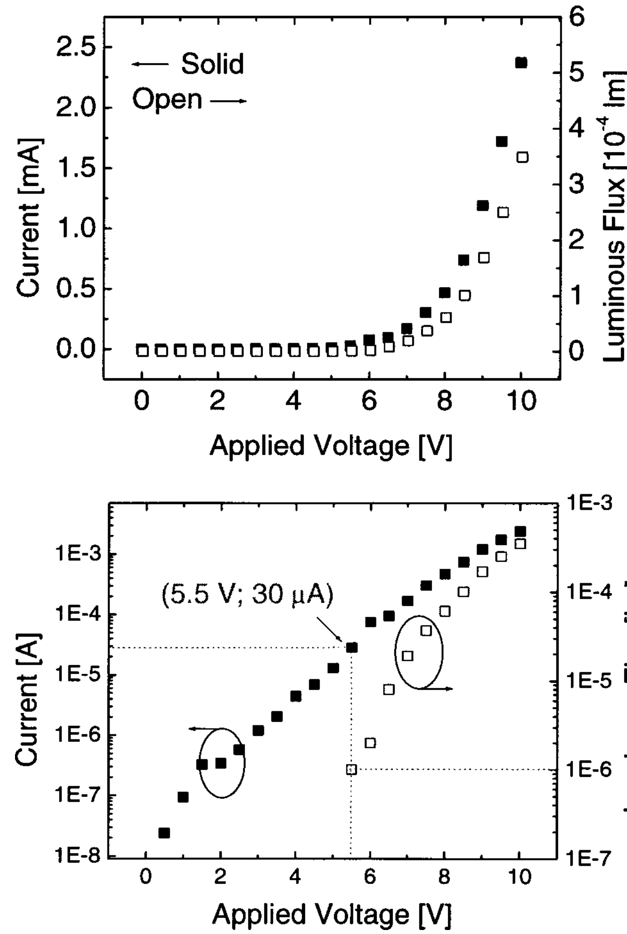

FIG. 5. Current and luminous flux vs applied voltage characteristics of pixel arrays of OPLEDs with ITO/PEDOT/red-emissive polymer/Al structure. The total pixel area is estimated to be 5 mm^2.

onds. We attribute this to the joule heating that occurs during device operations.[18] It has been shown previously that the temperature inside the device can reach such a level that it can cause intermixing of organic layers[19] and formation of nonemissive bubbles.[20] In our OPLED device, the insulation layer PMMA has a glass transition temperature of around 109 °C. Low T_g of PMMA might contribute to the contrast reduction for small pixels since the flow of the PMMA at high temperature and mixing of PMMA with other polymer layers could blur the pixel definition, or even eliminate the smallest ones as we observed for 2 μm size pixels. However, such a problem can be easily relieved by replacing PMMA with high T_g insulating polymers such as polycarbonate or thermal-curable polymers that can withstand much higher operation temperature after thermal curing.

The advantages of our OPLED fabrication technique are manifold. First, it avoids direct patterning of the emissive polymer and does not expose the electroluminescent polymer to high-energy radiation and alien chemicals during processing that can potentially compromise emission properties of the polymer. Second, since the imprint technique is a simple process and only requires inexpensive equipment, such a method offers low cost fabrication, which is pivotal for large-volume commercial applications. Third, this technique can be easily extended to nanoscale light-emitter fabrication. Once the mold is fabricated, it can be used repeatedly, which is very attractive for nanoscale feature patterns; other nanolithography techniques such as electron-beam lithography, on the other hand, involve very long writing time and very expensive equipment. In recent years, as micron-scale OPLED devices advance in sophistication, their remarkable properties raise intriguing possibilities if dimensions can be reduced to nanoscale. The imprint technique has demonstrated faithful patterning of sub-10 nm structures.[7] This high-resolution capability makes our pixel fabrication technique ideal for studying emissive properties of nanoscale OPLEDs.

IV. SUMMARY

An OPLED pixel fabrication method based on imprint lithography technology has been developed. We demonstrated light emissions from OPLED pixels on the order of several microns with this technique. This technique can be potentially extended to fabricate nanoscale OPLED light emitters.

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