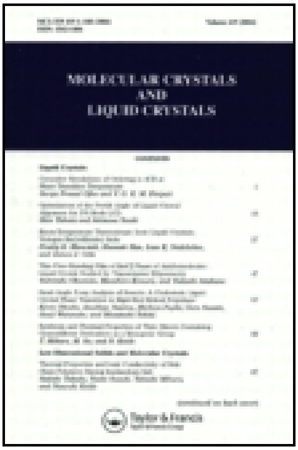

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Spontaneous Patterning of Polymer Film by Teflon Thin Film and Spin Coating for Polymer Light-Emitting Diode

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We have developed a selective coating technology for large-area polymer light-emitting devices (PLEDs). This can be done by using the different surface energy on the anode electrode. Teflon thin film can be used to make a hydrophobic surface (low surface energy) on the hydrophilic surface (high surface energy) of ITO for patterning of polymer film. Contact angle of ITO surface and Teflon coated one were 3 and 130°, respectively.

In order to demonstrate PLED by spontaneous patterning of polymer film we have fabricated PLED consist of 4 and 8 elements in series on the same substrate. The voltage of the devices required to achieve a particular brightness scales approximately with the number of elements in series.

Keywords: hydrophilic; hydrophobic; polymer light-emitting diode; polymer patterning; Teflon

INTRODUCTION

Manufacturing a large-area PLED with a single diode structure is difficult because the resistance of ITO electrode is high. Therefore, series connection of many small PLEDs on the same substrate is getting more attention for large area flexible white lighting [1]. Among various manufacturing issues needed to make large area lighting, the electrode contact between a cathode of one PLED and an anode of

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another PLED is not easy because hole transporting layer, such as a poly(3,4-ethylenedioxythiophene)-poly-(styrenesulfonate) (PEDOT:PSS), and emissive polymer layers can cover all cathode contact parts during spin coating process. The laser ablation was used to remove covering area by many companies [2,3]. However, this process has several demerits such as particle contamination and expensive for mass production.

When depositing material from a liquid, the surface energy of the substrate influences the amount and geometry of the deposited material. There are many approaches to make hydrophilic region on a hydrophobic surface or hydrophobic region on a hydrophilic surface for patterning of polymer films.

M. Böltau *et al.* described the micrometer size structures of polymer blends by micro contact printing (μCP) using poly(dimethylsiloxane) (PDMS) stamp [4]. H. Gau *et al.* reported the microchannels by hydrophilic stripes on a hydrophobic substrate [5].

Anton A. Darhuber *et al.* demonstrated the selective dip-coating of micropatterned surfaces with hydrophilic strips by photolithography [6]. The method reported above to pattern a surface is based on patterned surface modification by transfer of materials.

X. Wang *et al.* and J. Z. Wang *et al.* also exhibited patterning of polymer for application in organic electronics by dewetting [7,8].

In the present work we have developed a new patterning method for PLED device using a simple spin coating process and by a surface energy control. Teflon thin film is a polymer compound that is formed by chemical union of fluorine and carbon atoms. Teflon coating makes the surface from hydrophilic surface (high surface energy) to very hydrophobic (low surface energy) without any reduction in carrier injection [9].

Spontaneous patterning of water-soluble PEDOT:PSS and organic soluble polymers could be done on the patterned Teflon thin film because of its surface energy.

EXPERIMENTAL

The ITO substrates were cleaned by sonification in an isopropylalcohol (IPA), rinsing in deionized water, acetone and then irradiated by UV/Ozone before use. After surface cleaning of the substrate we made Teflon pattern on ITO layer by thermal evaporation with a shadow mask. The deposition rate of Teflon was 0.01 nm/s at base pressure of $<5 \times 10^{-8}$ Torr. The Teflon thickness 2 nm and the melting point of Teflon used in this study is 270°C.

For the polymer layer coating, PEDOT:PSS was spin-coated on the patterned Teflon substrates. These PEDOT:PSS coated samples were dried at 100°C for 10 min on a hot plate to remove water from the polymer layer. The polyfluorene-type blue polymer supplied by SK Corporation was dissolved in various solvents with 0.9 wt%. The polymer solution was spin-coated on the PEDOT:PSS coated substrates with 1000 rpm. This film was baked for 1 hour at 100°C. The contact angle was measured by the surface tension analyzer produced by Surface-Electro Cooperation.

RESULTS AND DISCUSSION

Spontaneous Patterning of Polymer Film by Spin-Coating

In order to characterize a change of material surface on the substrate we measured contact angle. Figure 1 shows the contact angles of the deionized water on various material surfaces. The contact angles of ITO and PEDOT:PSS on ITO are 3° and 6°, respectively. It means both surfaces are hydrophilic. The contact angle of ITO surface with Teflon of 2 nm is 130°. It indicates that ITO surface changed from hydrophilic to hydrophobic after deposition of Teflon.

Figure 2 exhibits the patterned PEDOT:PSS on the ITO substrate with stripe Teflon layer. Selective coating of water-soluble PEDOT:PSS could be done on the Teflon patterned ITO substrate because of totally different surface energy between the surface with/without Teflon layer. However, in case of hydrophobic emissive polymers, it is influenced by polymer solvent. Table 1 summarizes the characteristics of various solvents to use selective polymer patterning. The polarity index of these solvents is shown for comparison.

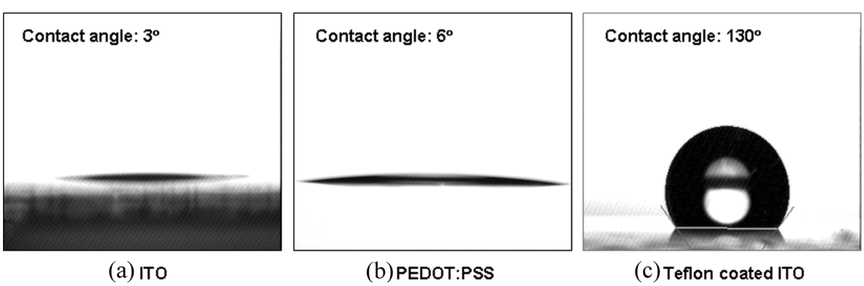

FIGURE 1 Contact angles of the deionized water on various material surfaces.

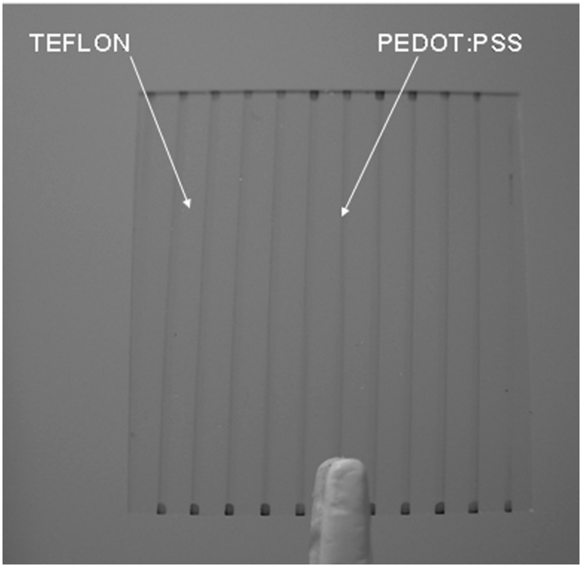

FIGURE 2 Patterned PEDOT:PSS on the ITO substrate with stripe Teflon layer.

We have investigated selective patterning of blue polymer on the patterned PEDOT:PSS substrate as shown in Figure 2. After making polymer solutions using 4 solvents, such as toluene, dichloroethane, tetrahydrofuran, and ethylacetate, we have spun the various polymer solvents to make thin film pattern on the substrate.

To verity the selective coating between the Teflon pattern and the PEDOT:PSS/Teflon one the samples put under the UV lamp of 365 nm. Figure 3 shows the optical images of the polymer films on the Teflon and PEDOT:PSS/Teflon patterned substrate.

TABLE 1 Characteristics of Various Solvents to Use Selective Polymer Patterning

	Polarity index	Boiling point (°C)	Viscosity (cP)
Toluene	2.4	111	0.59
Dichloroethane	3.5	84	0.79
Tetrahydrofuran	4.0	65	0.55
Ethylacetate	4.4	77	0.45
Water	9	100	1.0

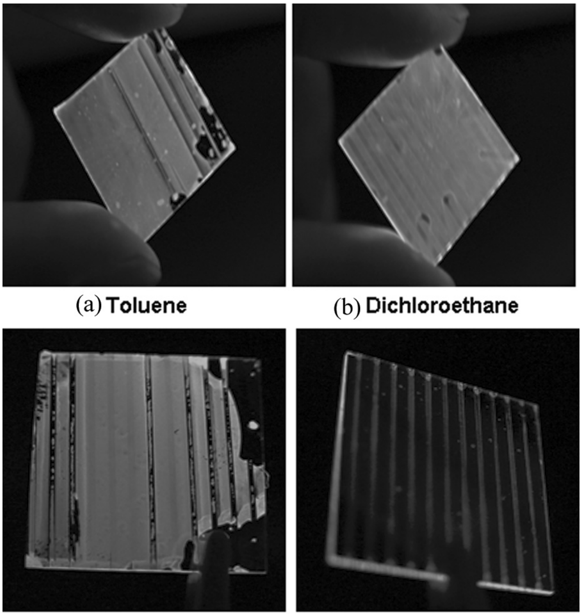

FIGURE 3 Optical images of the polymer films on the Teflon and PEDOT:PSS/Teflon patterned substrate under the UV-lamp of 365 nm.

As such a spontaneous patterning by spin coating the solvent polarity is a key process parameter. The polymer layers using solvents such as toluene, dichloroethane, and tetrahydrofuran were coated on the whole surface. The ethylacetate has higher polarity than the other solvents (toluene, and THF etc) as showed in Table 1. Therefore, using ethylacetate as a polar organic solvent for polymer coating solution we can achieve the selective polymer coating. The results of polymer patterning can be used for large area PLED for displays and lighting.

PLED Fabrication using Spontaneous Patterning

In order to demonstrate PLED with this technique, 4 elements and 8 elements series-connected PLED were fabricated on a $4\,cm \times 3\,cm$ substrate. The active area of each element of the 4 and 8 element devices was $20\,mm^2$.

Figure 4 shows the process procedure of PLED by spontaneous patterning of polymer layer: First process was photolithographic ITO patterning in wet station with ITO etchant. Second, photoresist

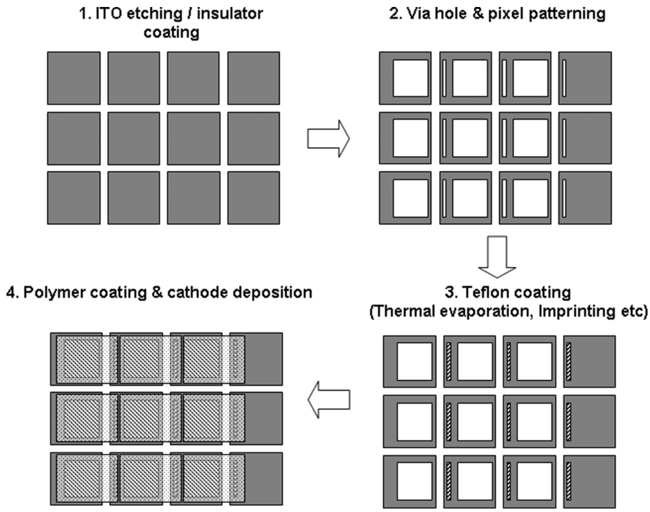

FIGURE 4 Process procedure of PLED by spontaneous patterning of polymer layer.

barrier for covering anode edge was formed by photolithography process. Third, Teflon layer of 2 nm was deposited by thermal evaporation with a metal shadow mask. Fourth, A selective polymer pattern was automatically formed by spin coating process because of strong hydrophobic surface of Teflon. Final process was the deposition of cathode metal.

To apply this architecture for a large size PLED using polymer solution it is important that the contact method, which is either between anode and cathode or between elements, in parallel without additional process such as laser ablation. In this study the contact parts between anode and cathode or between elements are comprised of ITO/Teflon/LiF/Al.

Figure 5 shows the semi-log current density–voltage curves as a function of the devices with different thickness of the Teflon layer. Since Teflon is an insulating material with an extremely high resistively of $10^{18}\,\Omega\,\mathrm{cm}$, it is easy to find that the current densities of the devices decrease with the increasing thickness of the Teflon layer. The resistance values of the device were 28.63 Ω for the device without Teflon layer, 31.82 Ω for Teflon thickness of 1 nm and 33.87 Ω for Teflon thickness of 2 nm, respectively. We found the resistance of device with Teflon layer slightly increased than that of the device without Teflon layer. But it is not critical point in this study

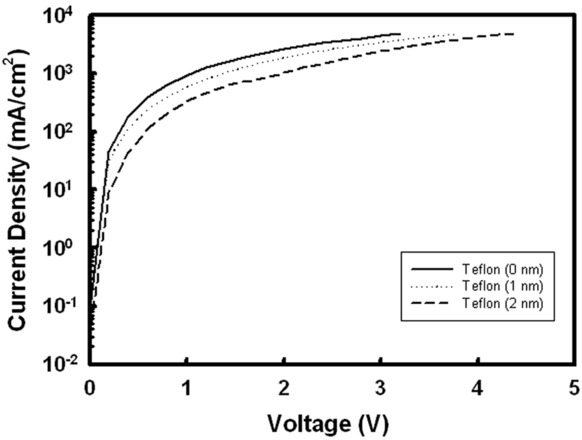

FIGURE 5 Semi-log current density–voltage curves as a function of the devices with different thickness of the Teflon layer.

because Teflon layer does not limit the injection of charge from ITO anode to cathode.

Figure 6 exhibits the electroluminescence intensity-voltage curves of devices with 4 and 8 elements in series on the same substrate. It compared with average data from a single element device. From Figure 6 we expected that the voltage of the devices required to achieve a particular brightness balances approximately with the value, which multiplies the voltage of single device by the number of elements in series.

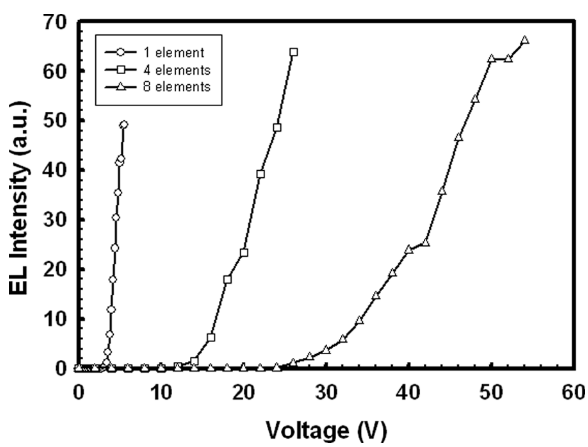

FIGURE 6 Electroluminescence intensity-voltage curves of devices with 4 and 8 elements in series on the same substrate.

CONCLUSION

We have introduced a self-organizing layer of polymer by spin coating on ITO anode. The Teflon layer of 2 nm was used for making a hydrophobic surface on the ITO layer. Using a Teflon layer we can achieve a selective coating of PEDOT:PSS and polymer emitting layer.

In order to demonstrate PLED by spontaneous patterning of polymer film we have fabricated 4 and 8 elements series-connected PLED. This technique should enable PLED applications such as backlight and lighting.

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