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Selectively modulated inkjet printing of highly conductive and transparent foldable polymer electrodes for flexible polymer light-emitting diode applications

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ABSTRACT

High efficient flexible polymer light-emitting devices which composed with highly conductive and transparent foldable polymer electrodes were fabricated. New doping materials, n-methyl-2-pyrrolidone (NMP) and n-methylformamide (NMF), have much improved the conductivity of poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS). The selectively modulated inkjet-printing method facilitated the PEDOT:PSS's application to both transparent anodes and highly conductive bus line electrodes. Multiple-time printed PEDOT:PSS electrodes showed a similar performance to Ag bus lines while one-time printed anode showed a much better figure of merit than that of ITO on plastic. Due to the flexible property, high transparency and high work function of the polymeric anode, ITO-free PLEDs showed a high performance with foldable characteristics.

1. Introduction

Recently, organic light emitting diodes (OLEDs) have been considered a front runner technology for transparent and flexible display applications, and have attracted a great deal of attention to implement novel future displays and areal lighting devices [1]. For realizing the transparent and flexible OLED displays, highly transparent, flexible, and conductive electrodes are essential. Although indium tin oxide (ITO) has been widely used as a transparent electrode material in the optoelectronic devices, it is difficult to be applied to the future optoelectronic devices with flexible features [2–8]. Moreover, for OLED applications, indium diffusion into the organic layer, especially hole injection layer (HIL), is one of the failure mechanisms [9,10]. The presence of indium in HIL increases the operating voltage and reduces the electroluminescence output efficiency [11]. In order to replace ITO, therefore, alternative transparent and conductive electrodes including silver nanowires [8,12–17], carbon nanotubes [18,19], thin metal films [20,21], graphene [6,7,22–25], and conductive polymers [26–28] have been intensively investigated. In particular, poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS) has emerged as one of the most promising materials for the transparent electrode in OLEDs and organic solar cells (OSCs) [29–36].

In fact, PEDOT:PSS is one of the well-known hole injecting material in the OLEDs due to its high transmittance and good electrical conductivity [37]. In addition to flexibility from the polymeric material property, PEDOT:PSS has an advantage of a facile controllability of its conductivity. Therefore, in order to use it as anode material, many research groups have doped PEDOT:PSS with various materials such as dimethyl sulfoxide (DMSO) [29],

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ethylene glycol (EG) [30,31], glycerol [32], and N,N-dimethylacetamide (DMAc) [33]. Although such anodes have been applied for OLEDs, their current and power efficiencies were still low comparing with the device with ITO anodes [29–35].

Another key advantage of the PEDOT:PSS is that patterning and thickness control of the thin film can be done by a simple inkjet printing process [34,35]. The usage of materials, the complicated photolithography steps, and the fabrication costs can also be reduced with the inkjet printing process. Since the PEDOT:PSS thin films have trade-off between transparency and conductivity depending on the film thickness, we can fully utilize these properties to implement both anode and bus line electrodes by just modulating the film thickness. The thickness of the PEDOT:PSS electrodes can be appropriately optimized for each usage by a simple inkjet printing method. Therefore, in this paper, we demonstrated the fabrication of highly efficient, flexible polymer light emitting diodes (PLEDs) with the inkjet-printed PEDOT:PSS anode and bus line electrodes.

2. Experimental methods

2.1. Preparation of substrates and fabrication of PLEDs

To prepare the polystyrene (PS, Sigma Aldrich, USA) coated polyarylite (PA, Ferrania, Italy) substrate (PS/PA), PS solution (1.8 wt% in toluene) was spun-coated on the PA substrate at 2000 rpm for 1 min, and the PS film was annealed at 120 °C for 30 min. The thickness of PS was 124.10 ± 5.60 nm. The schematic image of the structure of the fabricated PLEDs is illustrated in Fig. 1(a).

For comparison, we fabricated four types of PLEDs: PLEDs with ITO anode and bus line electrode on the glass (ITO-glass) or PA (ITO-PA) substrates: and PLEDs with PEDOT:PSS anode and PEDOT:PSS bus line electrodes (PEDOT-PS/PA) or Ag bus line electrodes (PEDOT-Ag-PS/PA) on the PS/PA substrates. The same organic layers of hole injection layer (HIL) and emissive layer (EML) and the cathode of LiF/Al bi-layer were used in all cases. Fig. 1(b) shows the fabrication process. The PEDOT:PSS anode was printed by the inkjet printer (Dimatix, USA) using a 10 pl cartridge. The drop speed was 8 m/s, and the inkjet printing process was conducted at the room temperature without heating. After the printing process, PEDOT:PSS films were annealed on the hot plate at 120 °C for 1 h. In the PEDOT-Ag devices, the Ag ink (Sigma Aldrich) was printed on the PEDOT:PSS anode with a 10 pl cartridge and with drop speed of 9 m/s. The printed Ag bus electrode was annealed on the oven at 120 °C for 20 min. In the PEDOT-PS/PA devices, the PEDOT:PSS bus electrodes were printed on the PEDOT:PSS anode by 7 times without a break. After 7 times printing, the PEDOT:PSS bus electrodes were annealed on the hot plate at 120 °C for 1 h.

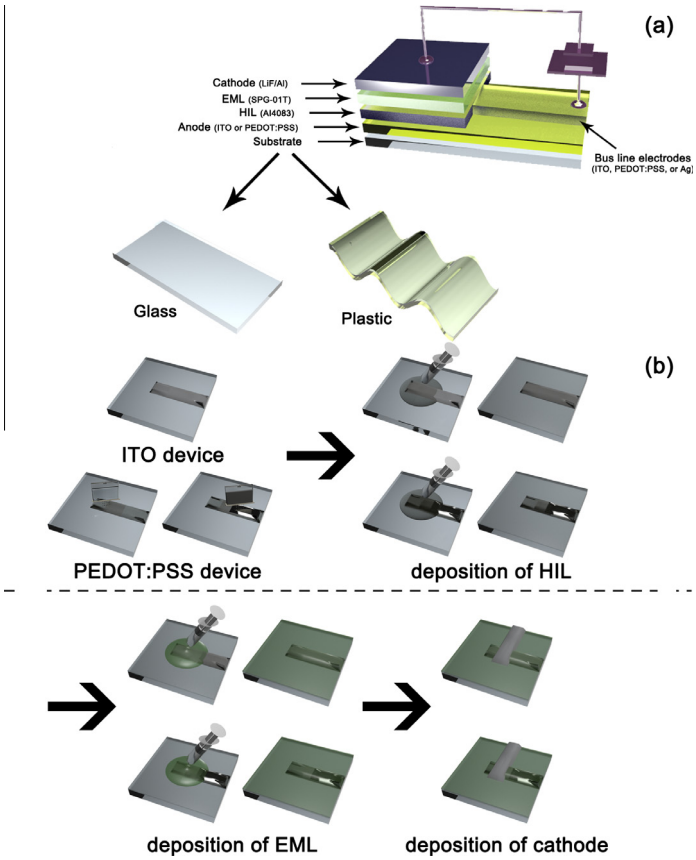

Fig. 1. (a) Structure of PLEDs (b) fabrication process.