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Highly efficient solution-processed inverted polymer light emitting diodes with uniformly coated poly(3,4-ethylenedioxythiophene):poly(styrene-sulfonate) layers on a hydrophobic emission layer using a dilution method

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ABSTRACT

Inverted polymer light emitting diodes (PLEDs), where solution-processed multi-layers were sandwiched between indium tin oxide and thermally evaporated aluminum electrodes, were fabricated and their characteristics were measured. In order to achieve high performance in the inverted PLEDs, there is a need for additional functional layers such as a hole transport layer and hole injection layer (HIL) on an emission layer (EML), but formation of the solution-processed layers on the hydrophobic EML have been rarely reported due to the extremely poor wetting property of a poly(3,4-ethylenedioxythiophene):poly(styrene-sulfonate) (PEDOT:PSS) solution, which is a widely used HIL. Non-wetting phenomena such as the formation of islands or radiation patterns in PEDOT:PSS films on the EML caused by their poor wetting property have been completely removed using a dilution method. By diluting PEDOT:PSS solutions with ethanol and analyzing a wetting envelope, we found the optimized dilution condition (10:1) and fabricated highly efficient inverted PLEDs with the uniformly coated PEDOT:PSS HIL, showing a current efficiency of 9.73 cd/A at 1000 cd/m^2 and uniform light emission simultaneously.

1. Introduction

Organic light emitting diodes/polymer light emitting diodes (OLEDs/PLEDs) have been considered as a promising technology for advanced displays with mechanical flexibility and optical transparency [1–7]. For commercial applications and reduction of fabrication costs, the PLEDs with a high efficiency and stability should be developed using a solution process such as a spin coating or printing process [8–19]. However, formation of the multi-layer structure by the solution process, which is essential to implement highly efficient and stable devices, is relatively challenging due to limited solvent orthogonality and thus intermixing between adjacent layers [20–25]. One of the promising approaches is adoption of an inverted structure, where anodes are located at the top of the whole devices. In general, the conventional PLEDs need vacuum-deposited low work function metals in cathodes for efficient electron injection, but most of solution-processed transparent or opaque electrodes have a high work function, which results in a low device efficiency due to a high electron injection barrier when they are applied to the cathodes [17,26]. In contrast, for the inverted structure, the electron injection can be enhanced by applying solution-processed functional layers on the cathodes. For example, air-stable zinc oxide (ZnO) has been deposited on the bottom cathodes using the spin coating as an electron transport layer to reduce the electron injection barrier [27–30]. In addition, the electron injection can be further enhanced by inserting an interlayer of polyethyleneimine (PEI) between the ZnO layer and emission layer (EML) because PEI can modify the work function of ZnO by inducing interface dipoles [31–33]. Unlike the feasibility of forming the solution-processed layers under the EML, additional coatings of functional layers on the EML by the solution process are hindered by its hydrophobic surface, making it challenging to obtain a solution-processed inverted PLED structure. Moreover, the introduction of a hole injection layer (HIL) on the EML is critical to achieving high performance in the inverted PLEDs with the ZnO/PEI layers because more efficient hole injection in the anodes facilitates more electron injection in the cathodes by hole accumulation at the PEI/EML interface [34].

To realize the high performance, most of the solution-processed inverted PLEDs have used a vacuum process for deposition of HILs such

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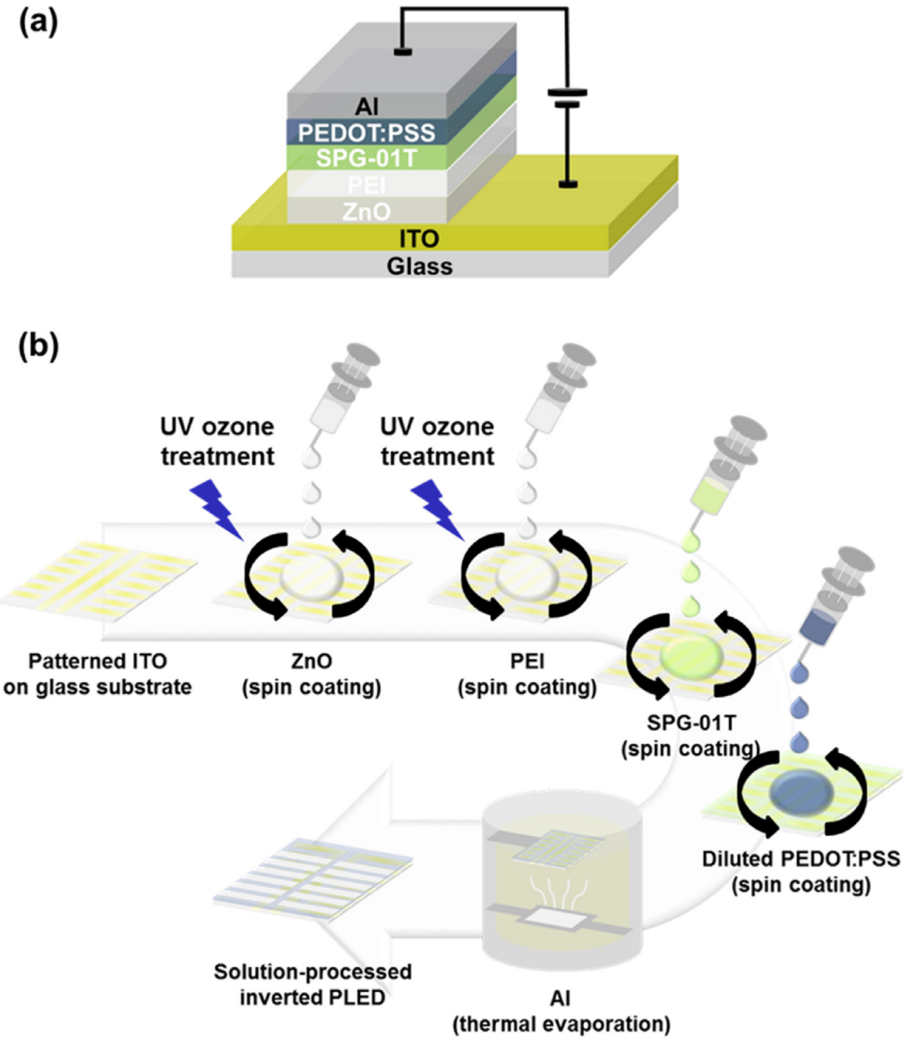

Fig. 1. (a) Device architecture of inverted PLEDs, (b) fabrication process.

as MoO_3 and top electrodes such as Au and Al on various hydrophobic EMLs [27,30,32,33]. In fact, several papers have been reported regarding the formation of poly(3,4-ethylenedioxythiophene):poly(styrene-sulfonate) (PEDOT:PSS) layers, which are widely used as the HIL in the PLEDs, on organic layers. In order to control the wetting property of PEDOT:PSS solutions on the generally hydrophobic organic layers, they used several approaches such as surfactant addition into the PEDOT:PSS solutions [35,36], surface treatment of the organic underlayers [37,38], and dilution of the PEDOT:PSS solutions with low surface tension liquids but only for organic solar cells [38,39]. It should be noted that the surfactant addition or surface treatment can affect the electrical, optical or morphological properties of the organic layers and thus degrade the device performance due to potential surfactant residue or chemical and physical transformation, respectively. On the contrary, the dilution method changes only surface tension of the original solution without causing any degradation of the organic layers, but still improving the wetting property. However, this approach has been rarely investigated in the PLEDs because the EMLs of the PLEDs show the more hydrophobic property than the organic layers of solar cells in general [40–43].

In this work, we demonstrated the inverted PLEDs with solution-processed multi-layers, especially the PEDOT:PSS HIL on the EML, sandwiched between indium tin oxide (ITO) and Al electrodes by modulating the wetting property. The poor wetting property of the PEDOT:PSS solutions on the hydrophobic EML was improved by diluting the PEDOT:PSS solutions with ethanol and the optimized dilution ratio was obtained by analyzing a wetting envelope of the EML and corresponding the surface tension for the diluted solutions. The fabricated devices showed not only the high performance but also uniform light emission in one substrate. Based on our method, a systematical analysis for the wetting property enables the formation of uniform films by the solution process, which leads to high-performance and low-cost electronic devices.

2. Experimental methods

2.1. Fabrication of inverted PLEDs

Inverted PLEDs were fabricated on glass substrates with ITO electrodes which were used as cathodes in the inverted structures as shown in Fig. 1(a). The substrates were sequentially cleaned with acetone, isopropanol, and deionized water in an ultrasonic bath. After that, the cleaned substrates were stored in an oven set to 100 °C for 1 h to remove any moisture on the surface and then treated by UV ozone before the deposition of upper layers. The fabrication process is illustrated in Fig. 1(b). The ZnO solution was prepared with a method which was described by Y. Sun et al. [28]. Accordingly, zinc acetate dihydrate ($Zn(CH_3COO)_2{\cdot}2H_2O$, Sigma-Aldrich, 1 g) was dissolved in 2-methoxyethanol ($CH_3OCH_2CH_2OH$, Sigma-Aldrich, 10 mL) and ethanolamine ($NH_2CH_2CH_2OH$, Sigma-Aldrich, 0.28 g) was added to the solution for stabilization. The completed solution was vigorously stirred for 1 day in order to enhance hydrolysis reaction in air. The ZnO solution which was filtered with a 0.45 μm polytetrafluoroethylene filter was spin-coated at