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Multilayered phosphorescent polymer light-emitting diodes using a solution-processed n-doped electron transport layer

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ABSTRACT

Efficient multilayered green phosphorescent polymer light-emitting devices (PhPLEDs) were successfully fabricated using a solution-processed n-doped small molecular electron transporting layer (ETL) composed of 1,3,5-tris(N-phenyl-benzimidazol-2-yl)-benzene (TPBi) and CsF. We found that the electroluminescence properties of the devices with n-doped ETLs are significantly improved. The maximum luminance efficiency of the device with 7.5 wt% CsF doped TPBi ETL reached 26.9 cd/A, which is 1.5 times as large as that of the undoped device. The impedance spectra of the devices and electron transport properties of the CsF doped ETLs demonstrate that doping dramatically decreases the impedance and enhances the electrical conductivity. Similarly, enhanced performance of PhPLED is also observed by use of CsF-doped 4,7-diphenyl-1,10 -phenanthroline (BPhen) ETL. These results demonstrate that CsF can be used as an effective n-dopant in solution-processed devices.

1. Introduction

During the past few decades, polymer light-emitting devices (PLEDs) have been developed significantly owing to their potential applications in low-cost and large-area displays, lighting and flexible devices [1–3]. In particular, phosphorescent PLEDs (PhPLEDs) have attracted much attention because they can harvest both single and triplet excitons to achieve nearly 100% internal quantum efficiency [4]. Up to present, the performance of solution processed PhPLEDs trails that of the vacuum-evaporated phosphorescent devices based on small molecular emitting layers (EMLs) because thermal evaporated various functional layers can easily tune the balance of charge carriers in the EMLs while solution-processing is ruled by solvent limitations. In order to improve device performance, multilayered PhPLEDs have been developed by insertion of appropriate charge injection/transport layers at the cathode and anode interfaces. Solution-processed hole-injection materials such as poly(3,4-ethylenedioxythiophene): poly (styrenesulfonic acid) (PEDOT:PSS) [1,2,5], high work function metal oxides [6–9] and cross-linkable polymers [10–12] have been explored to act as the hole injection layers (HILs). However, vacuum-deposited low work function metals (Ca and Ba) [13], ultrathin alkali salts (CsF and Cs_2CO_3) [14] and small molecule electron transport materials such as 1,3,5-tris(N-phenyl-benzimidazol-2-yl)-benzene (TPBi), 4,7-diphenyl-1,10 -phenanthroline (BPhen) and 1,3,5-tri(m-pyrid-3-yl-phenyl)-benzene (TmPyPB) [15–17] are frequently used in these solution processed devices to enhance electron injection/transport, which lead to high cost and complexity of the devices [18]. Therefore, the multilayered PhPLEDs with the solution-processed electron injection/transport layer (ETL/ETL) are desirable to improve device performance.

Recently, solution-processable electron injection/transport materials with orthogonal solubilities compared to that of the EML have been developed to fabricate multilayered PLEDs [4,19–25]. For example, solution-processed n-type metal oxides (ZnO, TiO_2 and ZrO_2) are widely used as efficient EILs in the inverted PLEDs [26–29]. Various polyelectrolytes, which can be dissolved in polar solvents such as water and ethanol, have been employed as EILs to fabricate multilayered devices [4,23–25]. Additionally, new small-molecule electron-transport materials have been developed to fabricate highly efficient multilayered phosphorescent devices by orthogonal solution-processing [19,30,31]. For example, Earmme et al. developed a small-molecule electron-transport material 1,3,5-tris(4-phenylquinolin-2-yl)benzene with a wide bandgap of 3.4 eV for the solution-processed multylayered blue PhPLEDs

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based on bis(3,5-difluoro-2-(2-pyridyl)-phenyl-(2-carboxypyridyl) iridium(III), giving a maximum external quantum efficiency (EQE) of 15.5% [19]. Jianget al. reported an alcohol-soluble electron-transport small molecule tris[4-(diphenylphosphoryl)phenyl]benzene for fully solution-processed multilayered white phosphorescent devices which achieved a maximum EQE of 11.5% [30]. Moreover, several commercial electron-transport materials, such as BPhen, TmPyPb and 3-(4-biphenyl)−4-phenyl-5-(4-tert-butylphenyl)−1,2,4-triazole (TAZ), have been used as ETLs solution-deposited from formic acid or methanol-based solutions for multilayered PhPLEDs [20,32,33]. However, a vacuum-deposited ultrathin alkali salt buffer layer (like CsF and LiF) is usually inserted between solution-processed ETL and cathode to reduce electron-injection barrier. As an alternative approach, n-doping can enhance electron-injection and transport of ETLs. The n-doping of ETL is typically realized by co-evaporating the electron-transport materials with a strong electron donor like Cs_2CO_3, LiF and LiN_3 [22]. Very recently, n-doping of small molecule electron transport materials by alkali carbonates (Cs_2CO_3 and Li_2CO_3) have been developed by solution-processing [21]. It has been reported that, the n-doped ETLs show considerable reduction in driving voltage and improvement in device efficiency compared to a conventional device [22].

In this work, we present efficient multilayer PhPLEDs using CsF as an effective n-dopant for solution-processed ETLs. We found that the solution-processed n-doped ETLs exhibited smooth surface morphology. Incorporation of the dopant into TPBi and BPhen significantly improved the performances of the devices. The devices exhibit high current efficiencies of 26.9 cd/A (CsF doped TPBi) and 20.8 cd/A (CsF doped BPhen), which are 50% and 72% higher than that of the reference device with undoped ETLs. The impedance spectra of the devices and electron transport properties of the CsF doped ETLs manifest that doping dramatically decreases the impedance and enhances the electrical conductivity.

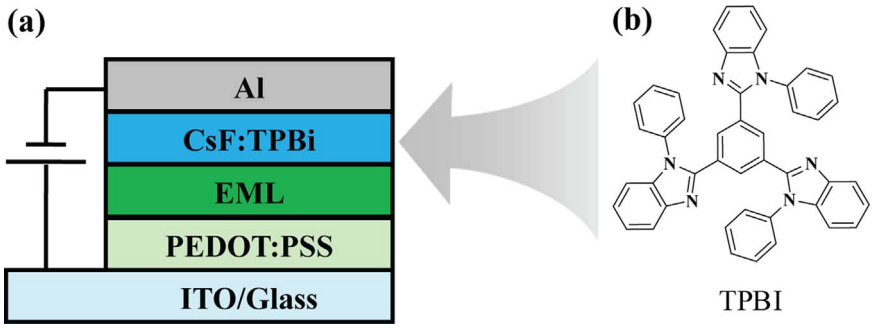

Fig. 1. Device configuration and molecular structure of TPBi used in this study.

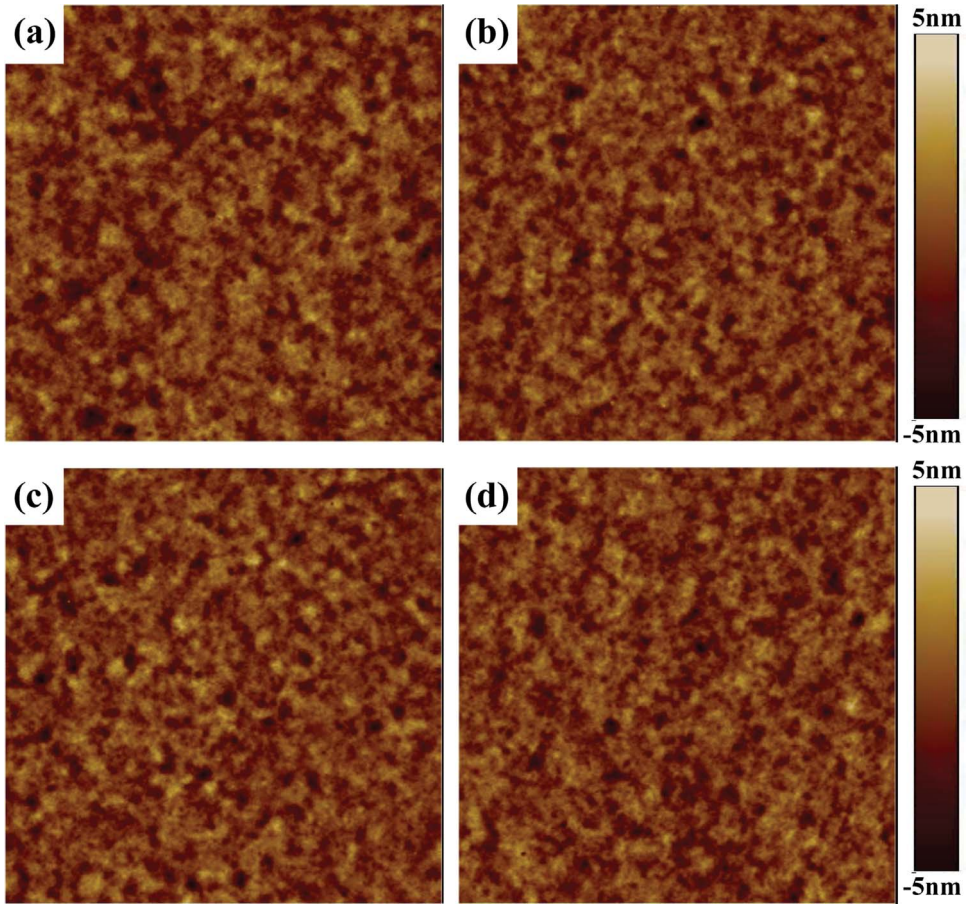

Fig. 2. Tapping mode AFM topographic images (5×5 μm^2) of (a) TPBi, (b) 5.0 wt% CsF:TPBi, (c) 7.5 wt% CsF:TPBi, and (d) 10.0 wt% CsF:TPBi.