



Phosphorescent dye-doped hole transporting layer for organic light-emitting diodes



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ABSTRACT

Hole transport materials are critical to the performance of organic light-emitting diodes (OLEDs). While 1,1-bis(di-4-tolylaminophenyl)cyclohexane (TAPC) with a high triplet energy is widely used for high efficiency phosphorescent OLEDs, devices using TAPC as a hole transport layer (HTL) have a short operating lifetime due to the build-up of trapped charges at the TAPC/emitting layer (EML) interface during device operation. In this work, to solve the operating stability problem, instead of using conventional HTLs, we use a *fac*-tris(2-phenylpyridine)iridium (III)(Ir(ppy)₃) doped layer as an HTL to replace the conventional HTLs. Because of the hole injecting and transporting abilities of the phosphorescent dye, holes can be directly injected into the emitting layer without an injection barrier. OLEDs based on a phosphorescent dye-doped HTL show significant improvement in operational stability without loss of efficiency.

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1. Introduction

Hole transporting materials are used in multilayer organic light-emitting diodes (OLEDs) and they are critical to the device performance. Attributed to the presence of electron-donating nitrogen, aromatic amine derivatives have been extensively used for hole transporting materials in OLEDs [1–10]. Among them, *N,N'*-diphenyl-*N,N'*-bis(1-naphthyl)-1,1'-biphenyl-4,4'-diamine (NPB) has been widely used as a hole transporting layer (HTL) in fluorescent OLEDs due to its good hole transporting ability and good alignment of the highest occupied molecular orbital (HOMO) level with the anode work function [11–16]. However, using NPB in phosphorescent OLEDs is problematic because of its inherent low triplet energy (2.3 eV) relative to the dopants [17–19]. This problem is even more severe for blue emitting phosphorescent OLEDs because of the higher triplet energy of the emitter.

Another archetype hole transporting material based on arylamine derivatives is 1,1-bis(di-4-tolylaminophenyl)cyclohexane (TAPC). TAPC has also been extensively used for high efficiency phosphorescent OLEDs because of its high hole mobility and high triplet energy (2.87 eV) [20–25]. However, OLEDs based on TAPC HTL suffer from low operational stability originating from the build-up of trapped charges at the TAPC/emitting layer (EML) interface, leading to a voltage rise and luminance decay during device operation [26]. The short device lifetimes have severely limited the use of TAPC in OLEDs.

To realize phosphorescent OLEDs with good operation stability, the challenge is to identify a stable HTL with a high triplet energy. Various aromatic amine derivatives have been synthesized and none of them satisfy the requirements of high triplet energy and good stability for phosphorescent OLEDs. In this work, instead of using a conventional aromatic amine-based HTL in a phosphorescent OLED, it is replaced by a wide-gap organic host heavily doped with a phosphorescent dye. It has been reported that phosphorescence dopants such as *fac*-tris(2-phenylpyridine)iridium

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(III) Ir(ppy)₃ is a good hole transport material [27,28] with a hole mobility of about 10⁻⁵ cm²/V s. By doping Ir(ppy)₃ into an organic matrix with a concentration exceeding the percolation limit, holes can be injected from the anode and transported via the dopant sites in the dye-doped HTL, and directly injected to the dopant sites in the emitting layer from the HTL without encountering any injection barriers as in the case of OLEDs with an aromatic amine HTL. There are two advantages with this approach. First, the hole injection barrier is completely eliminated because holes are injected from the same phosphorescent dye molecule. Second, quenching due to the low triplet energy in the HTL at the dye-doped HTL/EML interface is avoided as long as the triplet energy of the host material in the HTL is large. That is not a problem because hole transport is via the dopant sites and a large bandgap inert host can be used. With this strategy, we expect the device operating lifetime can be significantly improved. Herein, we investigate the carrier injection and transport characteristics of a phosphorescent dye and their effect on operational stability and current efficiency of OLEDs.

2. Experimental

Materials, device fabrication and characterization: OLEDs are fabricated using poly(3,4-ethylenedioxythiophene):poly(styrene sulfonate) (PEDOT:PSS) as the hole injection layer (HIL), TAPC as the HTL, 4,4'-bis(*N*-carbazolyl)biphenyl (CBP) as the host, and 1,3,5-tris(2-*N*-phenylbenzimidazolyl)benzene (TPBi) as the host as well as the hole blocking layer, Ir(ppy)₃ as the dye dopant, tris(8-hydroxyquinoline)aluminum (Alq₃) as the electron transporting layer (ETL), and lithium fluoride (LiF) as the electron injection layers (EIL). OLEDs in this study were prepared on patterned ITO glass substrates. Before fabrication, the substrates were rinsed by sonication in acetone and isopropyl alcohol, followed by a 15 min UV-ozone treatment. A 30 nm-thick PEDOT:PSS layer was spin-coated onto the substrates followed by thermal annealing at 140 °C for 30 min. All small molecules and cathode electrodes were evaporated by vacuum thermal deposition under a vacuum pressure of 3.0 × 10⁻⁶ Torr. All organic layers were evaporated at a rate of 1.0–2.0 Å/s, and LiF and aluminum (Al) were evaporated at a rate of 0.1 and 2.0–3.0 Å/s, respectively. The active area of the devices was 4 mm². The devices were encapsulated with a cap glass by using an UV-curable epoxy in a nitrogen (N₂)-filled glove box. The structure of hole only devices (HODs) for the measurement of hole mobility is ITO/PEDOT:PSS/TAPC (300 nm) or TPBi:30% Ir(ppy)₃ (300 nm)/MoO₃ (5 nm)/Al (100 nm) [29,30]. Another device structure to investigate the effect of Ir(ppy)₃ doping concentration on hole transport in TPBi:Ir(ppy)₃ layers is ITO/PEDOT:PSS/TAPC/TPBi:*x* % Ir(ppy)₃ (80 nm, *x* = 0, 8 and 30)/MoO₃(5 nm)/Al (100 nm). The structure of the OLED based on EML of 30% Ir(ppy)₃ doped TPBi layer is ITO/PEDOT:PSS/TAPC (40 nm)/TPBi:30% Ir(ppy)₃ (30 nm)/TPBi (10 nm)/Alq₃ (30 nm)/LiF (1 nm)/Al (100 nm). Electrical characterizations were performed using a Keithley 2400 source meter and the luminance was measured using a Konica-Minolta

LS 100 luminance meter with a close-up lens. All measurements were carried out in ambient atmosphere and room temperature.

3. Results and discussion

The device structures and the energy levels [31–33] of the molecules used in the devices are shown in Fig. 1. Three types of phosphorescent OLED devices were fabricated in this work (detailed fabrication process is described in Experimental Section):

- Device A: ITO/PEDOT:PSS/TAPC (40 nm)/CBP:8%Ir(ppy)₃ (30 nm)/TPBi (10 nm)/Alq₃ (30 nm)/LiF (1 nm)/Al (100 nm).
- Device B: ITO/PEDOT:PSS/Ir(ppy)₃ (10 nm)/ TPBi:30% Ir(ppy)₃ (30 nm)/TPBi:8%Ir(ppy)₃ (30 nm)/TPBi (10 nm)/Alq₃ (30 nm)/LiF (1 nm)/Al (100 nm).
- Device C: ITO/PEDOT:PSS/TAPC (20 nm)/TPBi:30% Ir(ppy)₃ (20 nm)/TPBi:8% Ir(ppy)₃ (30 nm)/TPBi (10 nm)/Alq₃ (30 nm)/LiF (1 nm)/Al (100 nm).

Here, Ir(ppy)₃ is used as a hole transporter in the TPBi matrix since pristine Ir(ppy)₃ has a high hole mobility (10⁻⁶–10⁻⁵ cm²/V s) which is comparable to the hole mobility of amorphous hole transporting materials such as NPB and CBP (~10⁻⁵ cm²/V s) [27,28]. By doping by using the TPBi layer with 30% Ir(ppy)₃, it is expected the dye-doped TPBi layer functions as an HTL since a doping concentration of 30% is well above the percolation limit. In this work, Device A is a reference device with TAPC as an HTL and Ir(ppy)₃-doped CBP as an emitter [34–36]. Device B has a PEDOT:PSS/Ir(ppy)₃ layer as an HIL and the TPBi:30% Ir(ppy)₃ as an HTL. Device C has a TAPC/TPBi:30% Ir(ppy)₃ layer as an HTL such that the dye-doped HTL is in contact with the EML and the TAPC layer is not in contact with the EML. In Devices B and C, TPBi is used as the host for the dye-doped HTL as well as the EML, and hole injection from the HTL to the EML is via the Ir(ppy)₃ hole transporter without encountering any hole injection barrier. It should be noted that TPBi is an electron transporter and it is expected that the Ir(ppy)₃-doped TPBi layer behaves as an ambipolar transporting emitting layer.

In order to compare the hole transporting capability of TAPC and 30% Ir(ppy)₃-doped TPBi layers, we fabricated the corresponding hole-only devices (HODs) and measured their electrical characteristics (the detailed structures of all HODs used in this study are described in the Experimental Section). Using these data, we obtained the space charge limited current (SCLC) hole mobility values of the TPBi:30% Ir(ppy)₃ and TAPC layers [37]. As shown in Fig. 2b and c, the hole mobility values of the TPBi:30% Ir(ppy)₃ and TAPC layers are 10⁻⁶–10⁻⁵ and 10⁻⁵–10⁻³ cm²/V s, respectively. In addition, we also investigated the effect of Ir(ppy)₃ doping concentration on the hole transporting properties. TPBi:Ir(ppy)₃ layers with different Ir(ppy)₃ doping concentrations (0%, 8% and 30%) on top of TAPC were fabricated. As shown in Fig. 2d, the hole current increases with increasing concentration of Ir(ppy)₃. These data show that the Ir(ppy)₃-doped TPBi layer does function as an HTL in Devices B and C.

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