Single-layer small molecular organic light emitting diodes without hole transport layer

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1. Introduction

Organic light emitting diodes (OLEDs) have received considerable attention for their potential application in flat panel display and lighting [1]. Typical structure of OLEDs based on small organic molecules are multiple organic layers sandwiched between the two electrodes, as layers for holes injection, holes transport, light emission, electrons transport, and electrons injection [2]. The multi-layer structure of OLEDs is fabricated by means of vacuum sublimation of organic molecules, and is too complicated for low-cost process of fabrication. In order to simplify the fabrication process, the single layer structure of small molecule OLEDs has been reported, in which the hole transport layer is replaced by an ultra-thin insulating layer such as copper oxide [3], self-assembled organosiloxanes [4], or a layer of conducting oxide, such as nickel oxide [5], or a layer of poly(3,4-ethylene-dioxithio-phene): poly(styrene-sulfonate) [6]. Our group also reported the high performance single-layer OLEDs with poly(tetrafluoroethylene) film [7,8]. However, further understanding of the mechanism of the single-layer molecule OLEDs is desirable.

In this paper, we developed three kinds of single-layer OLEDs based on tris-8-hydroxyquinoline aluminum (Alq3), in which Alq3 acted as the light emitting and electrons transport layer, whilst the ultra-thin insulating layer of silicon dioxide (SiO2), alumina (Al2O3) or polytetrafluoroethylene (Teflon™) instead of the hole transport layer were deposited between the anode and Alq3 layer respectively. The thickness of insulating layers were optimized, and all devices exhibited high performance. The hole injection of these devices was also investigated with hole-only devices. It was found that it is the hole tunneling effect and electrons blocking by the insulating layer that lead to the high performance of single-layer OLEDs.

2. Experimental details

The organic light emitting devices were built on glass substrates precoated with indium tin oxide (ITO) films with configuration of ITO/insulating layer (X nm)/Alq3 (60 nm)/LiF (0.8 nm)/Al (100 nm), where the insulating layer in the different devices were SiO2, Al2O3 or Teflon, respectively. The thickness of the insulating layers, X, was varied to determine the optimum thickness. The layers of SiO2, Al2O3 or Teflon were fabricated by alternating current sputtering, and Alq3, LiF and Al films were fabricated by thermal evaporation in a vacuum chamber. The deposition pressure during thermal evaporation was 1 × 10⁻³ Pa. The thickness of the films was determined in situ with a quartz-crystal sensor and ex situ by a profilometry (Nano-View MF-1000). The emission area of the device was 12 mm². The detailed process of devices fabrication can be found in our previous works [7-9].

The luminance–current–voltage (L–I–V) characteristics of the devices were measured by a computer controlled sourcemeter (Keithley 2602) and a calibrated silicon photodiode. All the measurements were carried out at room temperature under ambient conditions.

3. Experimental results and discussions

Fig. 1 shows the current density–voltage (J–V), luminance–voltage (L–V) and efficiency–voltage (E–V) characteristics of the single-layer...
device with SiO₂, Al₂O₃ and Teflon. As for the device with SiO₂ as the insulating layer, when the film of SiO₂ was 0.1 nm, the brightness of 2300 cd/m² was achieved on 7 V applied on the device, and the maximum of C–E was 1.8 cd/A. For the single-layer device with different thickness of Al₂O₃ film, the brightness was 2460 cd/m² at the 7 V applied on the device and 1.52 cd/A of C–E was achieved for the case of 0.1 nm-thick Al₂O₃ film. The Teflon film could be deposited by sputtering, which was quite smooth and had different surface morphology from Teflon films prepared by thermal evaporation [8]. When the Teflon film fabricated by sputtering was used as the insulating layer, the brightness of 5560 cd/m² was achieved on 15 V and maximum of C–E is 2.0 cd/A at the optimal film thickness of 0.7 nm.

In Fig. 1, according to the J–V characteristics of the single-layer devices with SiO₂, Al₂O₃ and Teflon layers, it was found that the current density of devices was reduced by the insertion of insulating layers. Since electrons dominate in the single-layer devices based on Alq₃, the attenuation of current density of devices were mainly resulted from the electrons blocking of these insulating layers.

We also investigated the holes injection of the single-layer devices using a “hole-only” device with the structure of ITO/insulating layer (X nm)/Alq₃ (60 nm)/N,N′-biphenyl-N,N′-bis(1-naphthyl)-(1,1′-biphenyl)-4,4′-diamine (NPB) (20 nm)/Ag (100 nm). Silver was chosen as the cathode with Fermi energy of 4.6 eV, and NPB was used as an electron blocking layer with lowest unoccupied molecular orbital (LUMO) of 1.8 eV. The great offset between the Fermi energy of the cathode and the LUMO level of NPB served to reduce the efficiency of the electron injection and to guarantee the holes injected from the anode to dominate in the device.

Fig. 2 shows the current density–voltage characteristics for the “hole-only” devices with insulating layers of SiO₂, Al₂O₃, or Teflon. It was found that the injected current density was enhanced by the insertion of the insulating layer and the maximum of current density of devices was achieved when the thickness of SiO₂, Al₂O₃, or Teflon films were respectively 0.1 nm, 0.1 nm and 0.7 nm. The enhanced hole injection was resulted from carriers tunneling effect, which have been studied in theory for the hole-only device in our previous works [9]. In this work, the relation between holes injection and performance of the single layer device were also shown in Fig. 3. Fig. 3 shows hole current density of hole-only devices and C–E of single layer devices for the cases of SiO₂, Al₂O₃, and Teflon layers with different thicknesses. It was shown that the enhancement of hole injection by insertion of insulating layer led to high C–E of single-layer devices.

For the single-layer devices based on Teflon layer fabricated by thermal evaporation, the maximum C–E was achieved with a Teflon layer of about 7 nm-thick [7]. This is different from that of a single-layer of sputtered Teflon, the optimal thickness being about 0.7 nm for the maximum of C–E. The discrepancy resulted from the different holes injection behavior. The rough surface and island pattern of Teflon film by thermal evaporation resulted in a maximum of hole tunneling injection with about 7 nm-thick Teflon film. However, because of the plane surface of Teflon film by sputtering, hole tunneling injection had the maximum when the Teflon film thickness
was about 0.7 nm [9]. Besides, the mechanism of holes injection enhancement and electrons blocking could be applied to the single-layer devices with the insulating layer of copper oxide [3], self-assembled organosiloxanes [4], poly(tetrafluoroethylene-perfluoroalkylylvinylethers) [7], and poly(tetrafluoroethylene-hexafluoropropylene) [7].

4. Conclusion

In conclusion, three kinds of high performance, small molecule, single-layer OLEDs based on Alq3 film with an anode buffer layer such as SiO2, Al2O3 or Teflon were developed and their characteristics were investigated. In the experiments, we found that the hole injection enhancement and electron blocking by the insulating layer in the single-layer devices led to the high performance of the single-layer OLEDs.

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