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

Zhaoxin Wu [a,*], Bo Jiao [a], Xuanke Zhao [a], Xun Hou [a], Liduo Wang [b], Haifeng Wang [b], Yudi Gao [b], Yong Qiu [b]

[a] Key Laboratory for Physical Electronics and Devices of the Ministry of Education, School of Electronic and Information Engineering, Xi'an Jiaotong University, Xi'an 710049, PR China
[b] Key Lab of Organic Optoelectronics & Molecular Engineering of the Ministry of Education, Department of Chemistry, Tsinghua University, Beijing 100084, PR China

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ABSTRACT

Organic light emitting devices based on tris-8-hydroxyquinoline aluminum with an ultra-thin insulating film instead of a hole-transporting layer were investigated. Three kinds of single-layer devices were fabricated with silicon dioxide (SiO_2), alumina (Al_2O_3) or polytetrafluoroethylene (Teflon™) as the insulating films, and they showed better performance than devices without insulating films. The hole injection of these devices was also investigated using hole-only devices. The holes injection enhancement by tunneling effect and electrons blocking of the ultra-thin insulating layers were experimentally proved to have yielded high performance of single-layer devices.

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-dioxythio-phene): poly(styrene-sulfonate) [6]. Our group also reported the high performance single-layer OLEDs with poly (tetrafluroethylene) 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 (Alq_3), in which Alq_3 acted as the light emitting and electrons transport layer, whilst the ultra-thin insulating layer of silicon dioxide (SiO_2), alumina (Al_2O_3) or polytetrafluoroethylene (Teflon™) instead of the hole transport layer were deposited between the anode and Alq_3 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)/Alq_3 (60 nm)/LiF (0.8 nm)/Al (100 nm), where the insulating layer in the different devices were SiO_2, Al_2O_3 or Teflon, respectively. The thickness of the insulating layers, X, was varied to determine the optimum thickness. The layers of SiO_2, Al_2O_3 or Teflon were fabricated by alternating current sputtering, and Alq_3, 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 mm^2. 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

* Corresponding author. Tel./fax: +86 29 82664867.
E-mail address: zhaoxinwu@mail.xjtu.edu.cn (Z. Wu).

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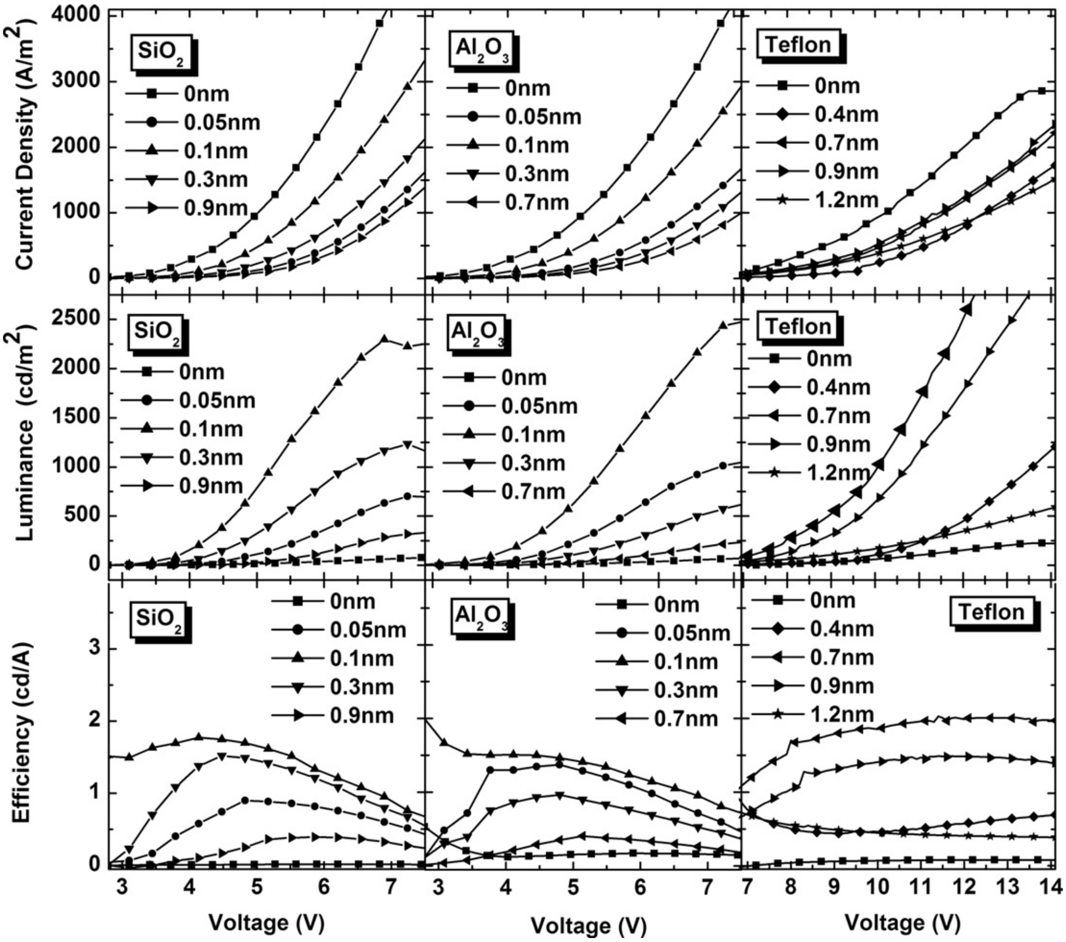

Fig. 1. Current density–voltage, luminance–voltage, and current efficiency–voltage characteristics of single layer OLEDs with SiO_2, Al_2O_3, and Teflon insulating layers of various thicknesses.

device with SiO_2, Al_2O_3 and Teflon. As for the device with SiO_2 as the insulating layer, when the film of SiO_2 was 0.1 nm, the brightness of 2300 cd/m^2 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 Al_2O_3 film, the brightness was 2460 cd/m^2 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 Al_2O_3 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/m^2 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 SiO_2, Al_2O_3 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 Alq_3, 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)/Alq_3 (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 SiO_2, Al_2O_3, 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 SiO_2, Al_2O_3, 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 SiO_2, Al_2O_3, 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 sputted 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

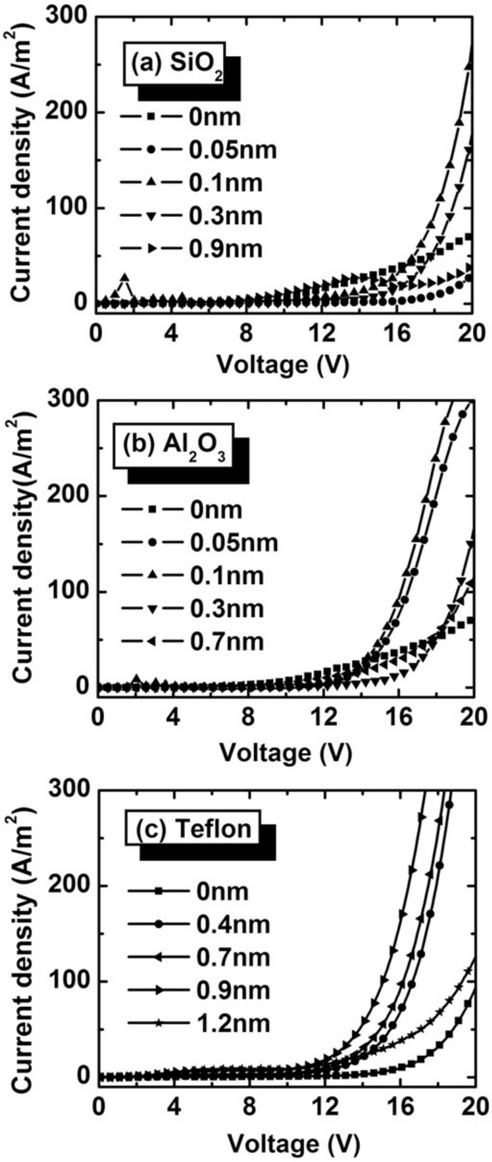

Fig. 2. Current density–voltage characteristics of a hole-only device with (a) SiO_2, (b) Al_2O_3, and (c) Teflon insulating layers of various thicknesses.

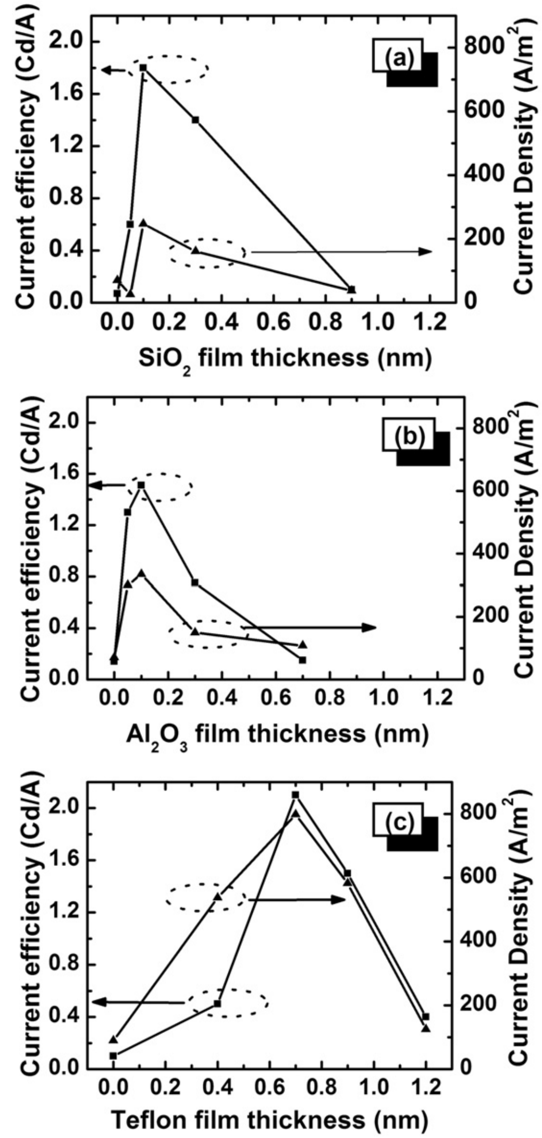

Fig. 3. Hole current density of "hole-only" devices (▲) and *C–E* of the single-layer OLEDs (■) using different insulating layers (SiO_2, Al_2O_3, and Teflon) with various thicknesses.

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-perfluoroalkylvinylethers) [7], and poly(tetrafluoroethylene-hexafluoropropylene) [7].

4. Conclusion

In conclusion, three kinds of high performance, small molecule, single-layer OLEDs based on Alq_3 film with an anode buffer layer such as SiO_2, Al_2O_3 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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