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Improved efficiency of organic light emitting devices using graphene oxide with optimized thickness as hole injection layer

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Abstract: Organic Light-Emitting Diodes (OLEDs) with graphene oxide (GO) as hole injection layer (HIL) have been demonstrated. The OLED devices possess structures of ITO/GO(x nm)/NPB(40 nm)/Alq_3(70 nm)/LiF(0.5 nm)/Al(100 nm), it is found that as the thickness of GO-HIL is a 3.6 nm an optimal current efficiency of 4.4 cd/A and a brightness of 15770cd/m^2 were achieved, respectively, which are higher than that of reference device without GO-HIL layer (1cd/A and 4735cd/m^2) . We reason that the improvement of electroluminescent (EL) intensity would be ascribed to the smoothed ITO surface and the reduced hole-injection barrier due to the high work function of GO layer. In terms of the impendence spectroscopy analysis of hole-only devices (HODs), for electrical character of device is mainly depends on the bulk resistance of the HODs. As a result, the optimized EL device offers fallen bulk resistances, finally, the improvement in EL performance was obviously realized.

Key words: Graphene oxide; Organic Light-Emitting Diodes (OLEDs); Hole only devices (HODs); Current efficiency; Impedance spectroscopy

1. Introduction

Organic light-emitting diodes (OLEDs) have considerably attracted attention due to their broad range of working temperature, low power consumption, wide viewing angle, fast response speed, high contrast, and vivid emissive color [1, 2]. In such devices various function layer, such as carrier injection and transporting materials, emitting materials and so on have been paid close attention and selected in device design [3, 4]. Furthermore, in order to enhance EL performance an interfacial layer was often inserted between anode and hole transporting layer (HTL). Such an interfacial layer is referred to hole injection layer (HIL). Anode/organic-interface of OLEDs plays importing role in increase EL performance because the HIL can reduce interface barrier of hole injection from anode to HTL so that better balance of electrons and holes into emitting layer (EML) can be realized. For this purpose, various interfacial layers have been inserted between the anode and HTLs, such as, poly (3,4-ethylenedioxythiophene):poly(styrene sulfonate (PEDOT:PSS) [5], graphene oxide (GO) [6],copper phthalocyanine (CuPC) [7], molybdenum oxide [8], tungsten oxide [9], and metal nanoparticles [10] have been employed. Among the HIL materials, the PEDOT: PSS is the most commonly used HIL material in OLEDs. However, a highly acidic aqueous solutions of PEDOT: PSS can corrode on ITO electrode [11] gradually and a significant quenching of radiative excitons [12] occurs between PEDOT:PSS and emission layer (EML) interface. Both will degrade the performance of devices eventually. In recent years, carbon-based two-dimensional (2-D) materials such as graphene [13-15] and graphene oxide (GO) [16-19] have been widely used in optoelectronic devices [20-23] due to their unique electrical and electronic properties. In particularly, graphene oxide is easily be dispersed in solvent and used in solution processed organic photonic devices because of the existence of the oxygen functional group [24]. Graphene oxide has a high work function and a large band gap, which is beneficial for hole injection in organic electronic devices [16, 25]. Graphene oxide has been introduced into the PEDOT:PSS and the PEDOT:PSS/GO composite films to be HIL [19] and anode [20], respectively. The graphene oxide treatment was respectively treated by oxygen- and hydrogen-plasma to act as HIL and HTL of the OLED devices. Particularly, Jesuraj et al. reported that as the oxygen treated graphene oxide was used as HIL an improved EL performance was affirmed. The EL current efficiency was enhanced from 3.3 cd/A to 4.2cd/A [6]. But Lee et al. have found that as the HTL was used in polymer LEDs the device performance is sensitive to GO layer thickness [17]. The function of GO HIL is described as improving hole injection rate. Note that GO also behaves as an insulator property due to its large band gap of around 3.6eV [16, 17]. So the GO thickness would play a crucial role in increase the performance of OLEDs.

In this manuscript, we report that the performance of OLEDs is strongly related to the thickness of GO-HIL between the ITO anode and NPB-HTL influenced by the surface morphology of GO coated ITO and the conductivity of the devices. In section 2, we describe the experiment. In section 3, we present the experimental results and give our discussions, including the surface root mean square (RMS) roughness of GO coated ITO, the device property of OLEDs, the conductivity of HODs and the resistance fitted through impedance spectroscopy. In section 4 we present a summary and conclusions.

2. Experiments

The indium tin oxide (ITO) glass substrate with a sheet resistance of about 5ohm/sq. was purchased by Shenzhen CSG Co., Ltd. Organic materials were purchased from Jilin Aolaide Co., Ltd. and Xi'an Bao Lite Optoelectronics

Technology Co., Ltd. All the materials were used without further purification. The GO ethanol dispersion was supplied by East China Normal University, GO nanosheets were synthesized from natural graphite by the modified Hummer's method [26]. The prepared a 0.60 mg/ml GO ethanol dispersion was added to ethanol for dilution. The concentration of GO used in the experiment in ethanol dispersion was controlled at 0.02 ~ 0.10 mg/mL.

Prior to the experiment, the quartz glass and ITO glass substrate were cleaned with a routine cleaning procedure [27]. Ultrasonic treatment was carried out on the ITO glass substrates in de-ionized water, ethanol and acetone for 15 min, respectively. Then the ITO substrates were dried in a vacuum oven and treated with oxygen plasma for 5 min to improve its polarity, adhesion and hydrophilic character [28]. The GO ethanol dispersion was spin-coated with a speed of 1000 rpm for 10 s and annealed at 120℃ for 15 min to remove the excess amount of solvent. Different thicknesses of GO layers ((a) x=0 nm, (b) x=1.8 nm, (c) x=2.4 nm, (d) x=3.2 nm, (e) x=3.6 nm, (f) x=4.0 nm, (h) x=4.8 nm, (g) x=5.4 nm) were spin coated with various concentrations of GO ethanol dispersion (0.02 mg/mL ~ 0.10 mg/mL). The evaporation of the organic thin film was carried out in an organic thermal evaporation system. The deposition rate is controlled at 0.02~0.04 nm/s under about 1×10^{-4} Pa base vacuum. Finally, 0.5 nm LiF and 100 nm of Al were evaporated under about 5×10^{-4} Pa base vacuum. The film thickness was monitored by the quartz crystal monitoring instrument. The absorption spectra of GO ethanol dispersion and GO attached on ITO surface were measured by an UV-vis spectrophotometer (U-3310), respectively. The roughness and surface morphology of the films were analyzed by an atomic force microscope (AFM) (Ntegra Spectra). The optoelectronic characteristics of the devices were measured by a Keithley 2400 Source Meter and a PR655 spectroradiometer. The impedance spectroscopy was measured with a Solartron 1260 impedance analyzer. A 0.1 V sine-signal voltage was applied on the sample with frequencies varying in steps from 0.1 to 10^6 Hz. All measurements were performed in an environment condition.

The device structure is shown in Fig.1 (a). GO is used as HIL, N,N'-Bis-(1-naphthalenyl)-N,N'-bis-phenyl-(1,1'-biphenyl)-4,4'-diamine(NPB) is used as HTL, Aluminum Tris(8-Hydroxyquinolinate) (Alq_3) is used as both light-emitting layer and electron transport layer. The energy level arrangement and the chemical structure of GO are shown in Fig. 1 (b) and (c), respectively. Here the device without GO layer is taken as the reference device.

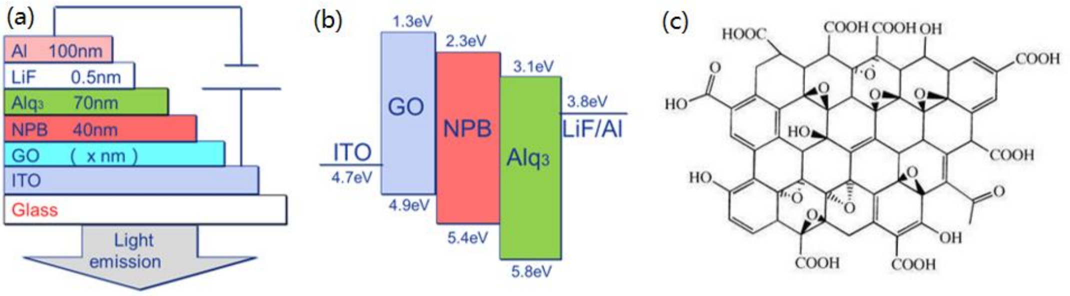

Fig.1 (a) Device structures of OLEDs with GO layer（b）Energy level diagram (c)The chemical structure of GO.

3. Results and discussion

The normalized UV-vis absorption spectra of GO ethanol dispersion (0.02 mg/ml) and GO layer (3.6 nm) on quartz glass are shown in Fig. 2. It can be seen that two GO films depict identical absorption peak. The UV-vis absorption peaks of different thicknesses of GO layers are similar to that of the GO layer with a thickness of 3.6 nm, while the absorption intensity is different. The max absorption peak of GO is located at 230 nm, which is assigned to $\pi \sim \pi^*$ transition of C=C double bond. Also there is a shoulder peak located at 300 nm, which is assigned to $n \sim \pi^*$ transition [6].

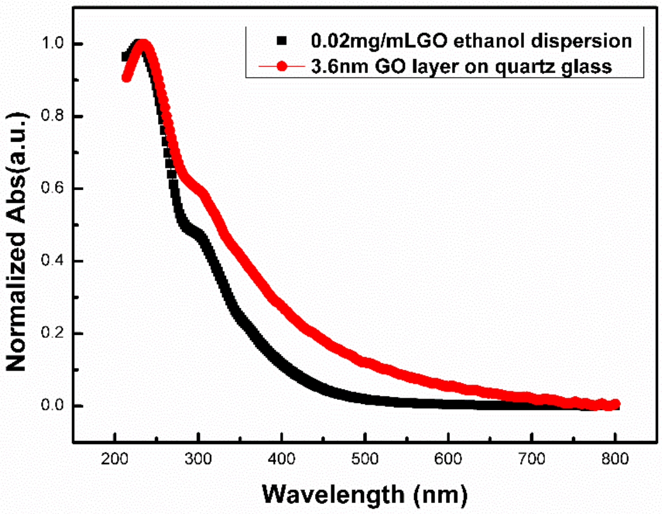

Fig. 2. The normalized UV-vis absorption spectra of GO ethanol dispersion and GO film on quartz glass.

It is well known that the surface morphology of the anode would influence OLED performance heavily because hole must inject from anode HTL into EML. If the anode possesses rough surface and higher surface resistance, such an anode based OLED device behaves falling EL properties, because the contact area between the anode and the organic layer will be smaller [20, 29]. Short behavior with cathode was taken even place as anode surface conductive layer is unduly rough. Here, the GO layer with different thicknesses was spin coated on patterned ITO surface. The morphology of bare ITO surface and GO coated ITO were studied by AFM, the AFM images and the surface roughness were shown in Fig. 3, indicating that the root mean square (RMS) roughness is 4.59 nm for bare ITO. For ITO surface with GO film the RMS varies with the GO film thickness. The RMS roughness decreased to 3.55 nm as 3.6 nm GO layer was used and increased again as the GO thickness became further thicker. When GO was spin-coated on the ITO glass, the GO-ethanol with interfacial wettability [24] will be filled into the defects of bare ITO conductive film, leading to an improved surface smoothness of ITO conductive film at first. However, when the thickness of GO layer was further increased, the redundant GO-dispersion would be stacked on the smooth surface so that the surface roughness raise. In this case, the GO film could be regarded as a buffer layer due to modification of the surface morphology of ITO anode. When 3.6 nm GO layer was used as HIL, there should be a better charge injection from the ITO anode to NPB-HTL because of presence of a better RMS roughness.

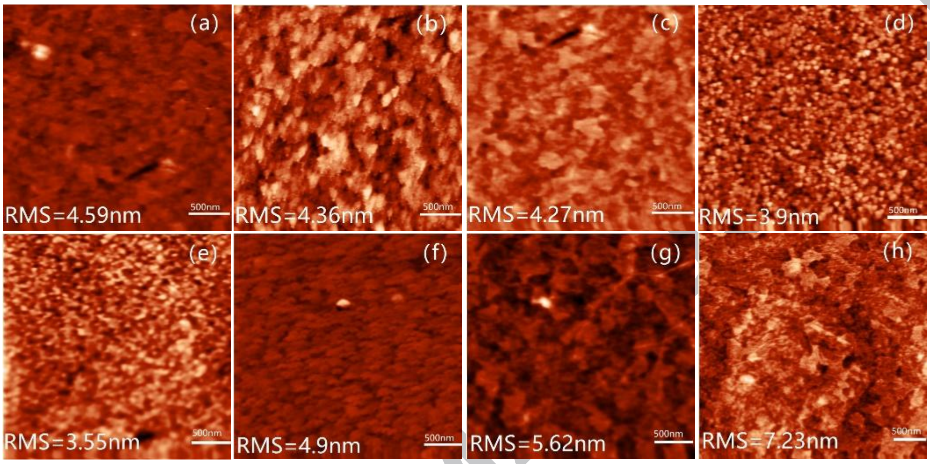

Fig. 3 AFM images of ITO / GO (x nm) here, (a) x=0 nm, (b) x=1.8 nm, (c) x=2.4 nm, (d) x=3.2nm, (e) x=3.6 nm, (f) x=4.0 nm, (h) x=4.8 nm, (g) x=5.4nm

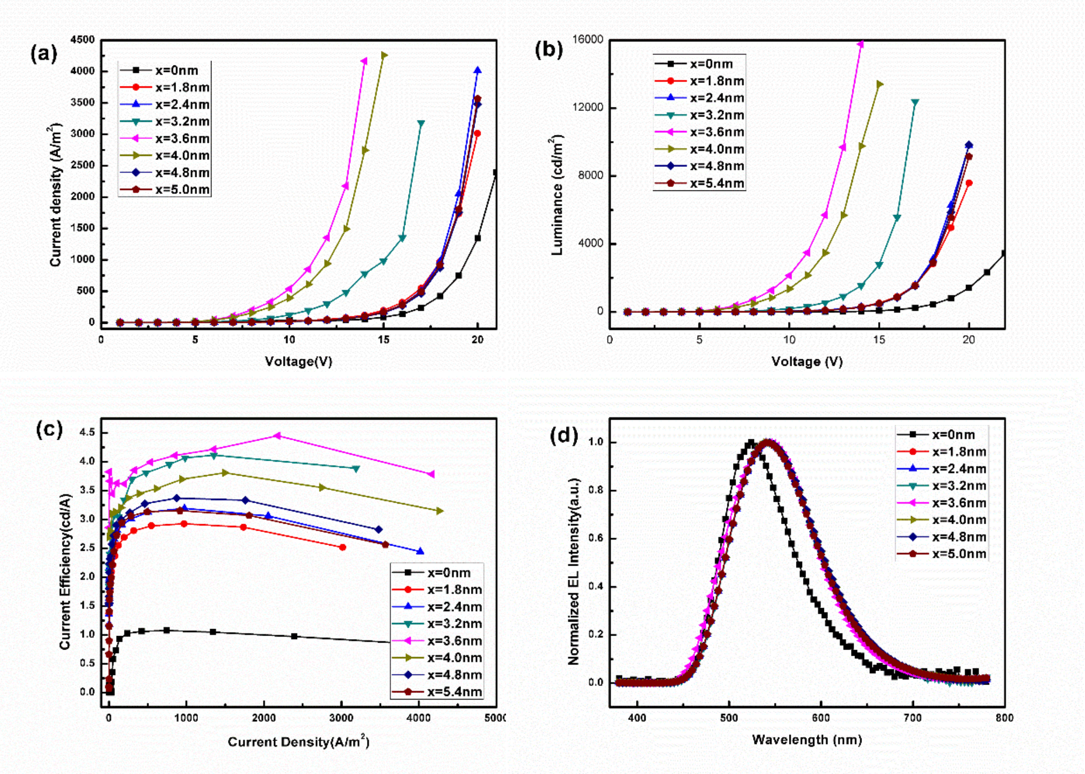

Fig.4 (a) Current density-voltage (b) luminance-voltage (c) current efficiency-current density characteristic (d) normalized EL spectra of the OLEDs.

The device characterizations and EL spectra of OLEDs with various thickness of GO HIL are presented in Fig. 4. It is observed that GO-HIL based OLEDs have rising EL intensity compared to the reference device. It implies that the GO-HIL effects on the electrical characteristics of the OLED devices heavily, as depicted in Fig. 4(a). The reference device exhibits only a current density of 39 A/m^2, but the device with 3.6 nm GO-HIL provides a max current density of 2178 A/m^2. At the same time, the turn-on voltage was reduced from 10 V to 4 V. The EL intensity of reference device is much lower than those of OLEDs with GO-HIL. The optimized OLED with a 3.6 nm thick GO-HIL exhibits a peak current efficiency of 4.4 cd/A and a highest luminance of 15770 cd/m^2, which is in striking contrast to the reference device. Based on the normalized EL spectra in Fig. 4(d), the EL peak of reference device lies at 524 nm, EL peak of GO-HIL lies at 541 nm, thus we can see that in contrast to EL peak of reference device red-shift EL peak of device with GO-HIL was determined.

As a result we can get that the thickness of GO-HIL could markedly influence on EL performance of the devices. The 3.6 nm GO-HIL based device exhibits a max EL intensity, which can be attributed to the improving hole injection rate, so that hole and electron in EML of this device would become balance. A higher hole injection rate from the anode to HTL is realized in two ways. Firstly, it is well known that GO has a higher work function than ITO. The electrons of ITO transfers to GO when GO inserted on ITO, then an interface dipole appears which will reduce the hole injection barrier in OLEDs [6]. Here, when GO layer is inserted between ITO and NPB, the hole injection barrier from ITO to NPB was reduced from 0.7 eV to 0.5 eV (as shown in Fig.1(b), which leads to the hole injection easily. Secondly, under Fig. 3(d), the morphology of ITO surface is improved for the sample with 3.6 nm GO layer. The lower surface roughness would result in a larger contact area between the ITO anode and the NPB-HTL which is benefit for the hole infection and lead to the increase of current density (J) and luminance (L), as shown in Fig. 4(a) and 4(b). To us all, the current efficiency is mainly dependent on the charge balance in the device [30]. In terms of the above discussed results, we consider that the holes injection is enhanced for the OLEDs with 3.6 nm GO layer, which could facilitates the carrier balance, leading to a higher current efficiency, as shown in Fig. 4(c). On the other hand, the hole-electron recombination zone is changed by the higher hole injection rate, which leads to a red-shift of the EL emission of device with GO-HIL [27]. All EL performances in this study are listed in Table 1.

Table 1 Device performances of OLEDs with different thicknesses of GO layer

Device(X nm)	0	1.8	2.4	3.2	3.6	4.0	4.8	5.4
Turn on voltage(V)	10	5	5	4	4	4	5	5
Maximum brightness(cd /m^2)	4735	7590	9810	12390	15770	13410	9837	9141
Maximum current efficiency (cd/A)	1.0	2.9	3.1	3.8	4.4	4.1	3.3	3.1

In general, the charge mobility is a crucial factor for clarifying carrier transporting ability. The conductivity variety of changed thickness GO layer based devices was confirmed by hole-only devices (HODs). The HODs with structures of ITO/GO(x nm)/NPB(40nm)/Al(100nm) were constructed. Fig. 5 exhibits the log J vs log V curves of HODs. We note that the current density of the HODs with GO layer is obviously higher than that of the reference HOD. The HOD with an optimal thickness of 3.6 nm GO layer provides the highest current density. In terms of the increase in current density due to the GO layer, it could be turned out that GO film on ITO is beneficial to the hole transport in HODs. According to the Mott-Gurney space-charge-limited-current (SCLC) model [31, 32] reported in Refs. 31 and 32, the hole mobility in HODs can be fitted, as bellow:

$$J_{SCLC} = \frac{9}{8}\varepsilon_r\varepsilon_0\mu_h\frac{V^2}{d^3}$$

Here, [$\varepsilon_0 = 8.85\times10^{-14}$C/V· cm] denotes the dielectric constant of vacuum, ε_r is the relative permittivity of the active layer material [33], d is the thickness of the active layer, and μ is the carrier mobility. The hole mobility of HODs with different thickness of GO layer are shown in Fig. 6. The hole mobility of the HOD with an optimal thick GO-HIL of 3.6 nm is calculated to be 1.7×10^{-8} cm^2/Vs, which is improved by two orders of magnitude compared to that of reference HOD (3.4×10^{-10} cm^2/Vs). With increasing thickness of GO layer, the hole mobility increases up to a max value at 3.6 nm and then lessen. The quantitative comparison of the hole mobility of these HODs confirmed that the use of GO-HIL promoted hole transport. The increase in conductivity of the OLED device leads to a balance of carrier transporting. The highest current efficiency of 4.4 cd/A and a luminance of 15770 cd/m^2 are obtained from the devices with optimal thick GO layer.

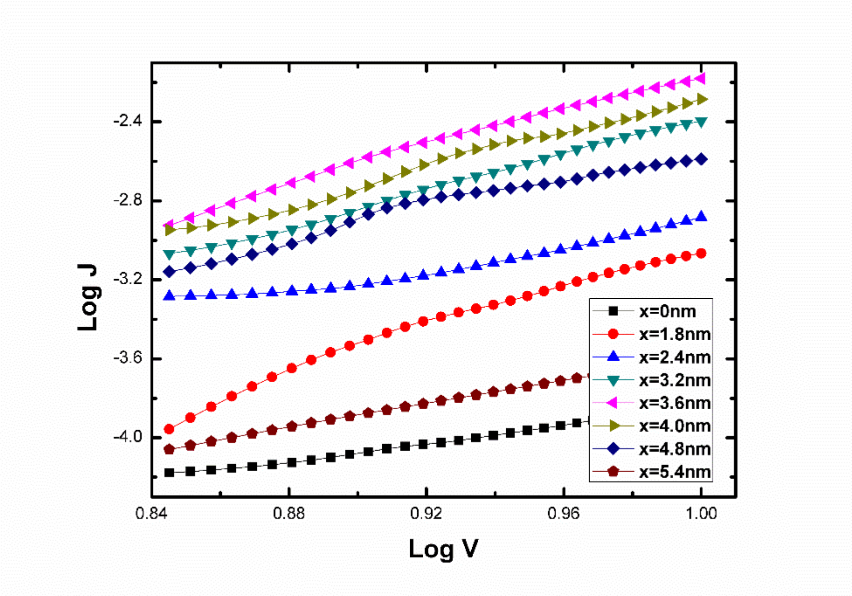

Fig.5 Log J vs log V curves of HODs with different thicknesses of GO layer.

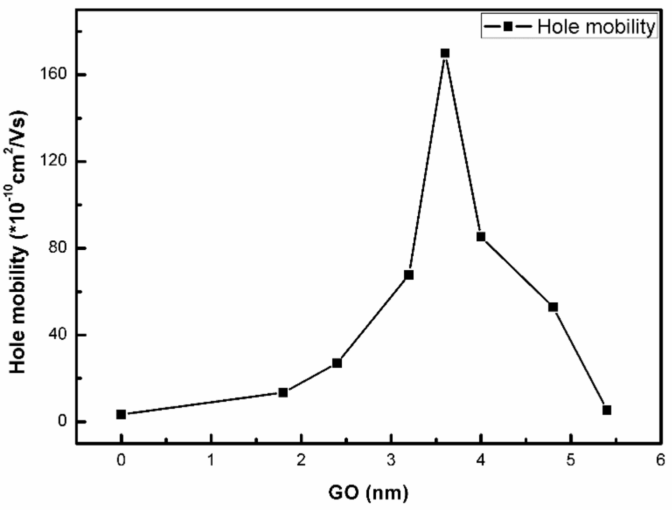

Fig.6 The calculated hole mobility of HODs using the Mott-Gurney Law.

According to Ref. 34, the impedance spectroscopy was measured and the Nyquist plots of the impedance spectra of HODs with various thickness GO layer were provided, as shown in Fig. 7, which also support that device with 3.6 nm thickness GO layer offers the highest efficiency. An equivalent circuit (Fig. 8 inset) utilized in the spectral analysis consists of two *RC* subcircuits in series, each of which are respectively attributed to the interface resistance (R_i) and bulk resistance (R_b) contributions. The capacitors are represented by constant phase elements (CPEs) with their impedance given by $Z_{CPE}=B^{-1}(j\omega)^{-n}$, where B is a constant that is independent of frequency. The simulation of the impedance spectra at 3.6 nm based on the equivalent circuit, as an example, is given in Fig. 8. The high correlation indicates that the proposed equivalent circuit accurately reflects the experimental data. Fig. 9 exhibits the bulk resistance, interface resistance and total resistance of HODs with different thick GO layer. It shows that there is only a high bulk resistance and absence of interface resistance in the reference HOD, which means an ohmic contact between the ITO film and NPB-HTL. When GO layer is inserted between ITO film and NPB layer, the bulk resistance of HODs reduced and the interface resistance between the ITO film and NPB layer appeared. The reduced bulk resistance leads to a higher hole mobility, providing a higher current density at the same voltage, as shown in Fig.6. The lowest bulk resistance, interface resistance and total resistance appear in the HODs with 3.6 nm thick GO layer, which leads to the highest current efficiency and the best performance of OLEDs.

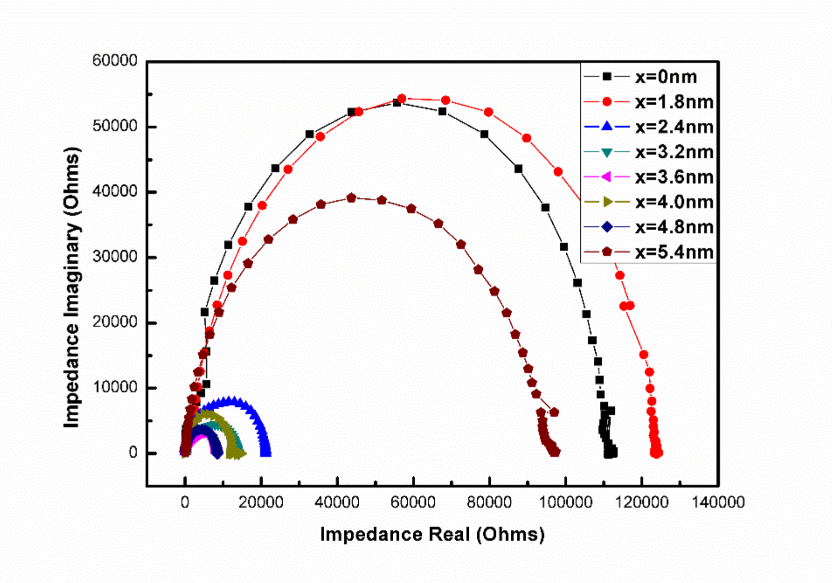

Fig.7 Nyquist plots of HODs with different thicknesses of GO layers.

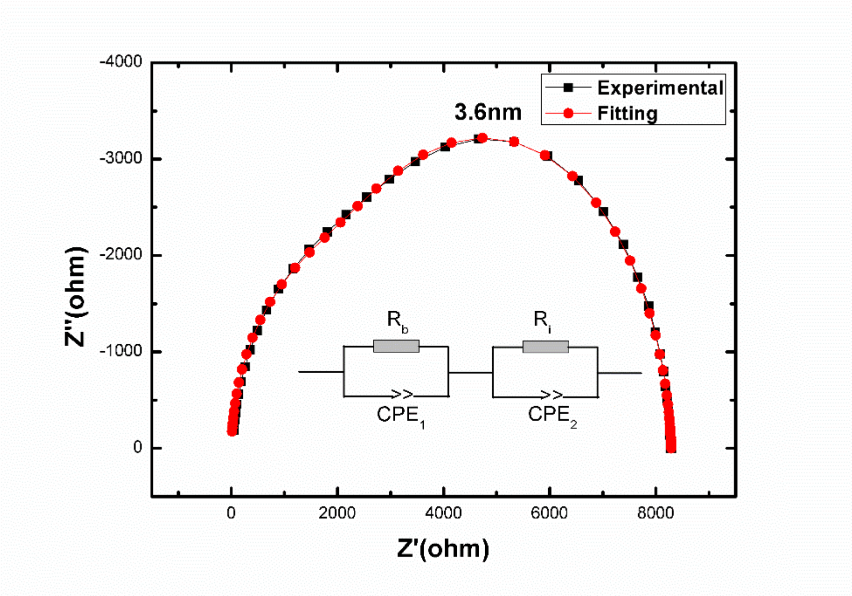

Fig. 8. Simulation of the impedance spectra of HODs with 3.6 nm thickness of

GO layer by the series *R-CPE* equivalent circuit shown in the inset. Solid circle is experimental data and the solid line is the fitted curve based on the equivalent circuit.

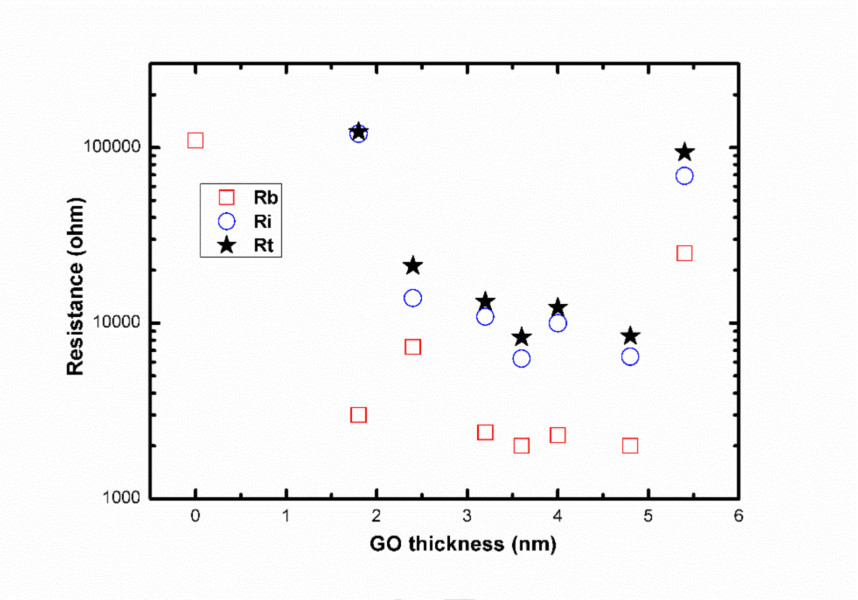

Fig. 9 The bulk resistance (R_b), interface resistance (R_i), and total resistance (R_t) of HODs with different thicknesses of GO layer.

As discussed above, the EL performance with all devices with GO-HIL are better than reference device. The GO layer acting as an inset reduces the hole injection barrier and falls roughness of ITO surface, resulting in an increase in hole injection rate. It should be noted that the GO layer also behaves as an insulator because of its broad energy band gap. When the thickness of GO layer increases further, both the interface-and bulk-resistances were increased. The higher interface resistance could hindrance to the carrier injection. Both the increased bulk-and interface-resistance would lower current density. When an optimal thick GO layer of 3.6 nm was used, the HODs showed the lowest two resistances, and the OLED exhibited a max current efficiency of 4.4 cd/A and a peak brightness of 15770 cd/m^2.

4. Conclusion

The effects of GO-HIL layer on the performance of OLEDs were investigated in this work. OLEDs with improved efficiency were prepared by utilizing different thicknesses of GO-HIL. An optimal device with a 3.6 nm GO-HIL provides a max brightness of 15770 cd/m^2 and a max current efficiency of 4.4 cd/A. Furthermore, in order to explain the mechanism of demonstration of such a high EL intensity, we determined the RMS roughness of GO coated ITO, the current density-voltage curves as well the impedance spectroscopy of HODs. We observed that the improvement should be ascribed to improvement of surface roughness of ITO anode, the reduction of bulk resistance and the increase of hole mobility. We obtained that inset of GO-HIL between ITO and NPB-HTL offers favorable effect. We believe that the positive influence would be used in the design of another organic electronics devices.

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The effects of GO-HIL layer on the performance of OLEDs were investigated in this work, the highlights of this paper are as follows:

1. The effect of GO thickness on the performance of OLED devices used as HIL was studied in this manuscript. An optimal device with GO-HIL the optimal thickness of 3.6nm provides the maximum brightness and current efficiency, which is 3.3 times and 4.4 times higher than that of reference OLED$_S$, respectively.
2. Based on the impedance spectroscopy analysis of hole-only devices (HOD$_S$), the electrical character is mainly depends on the bulk resistance of HOD$_S$.

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