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Significantly Enhancing Luminance of Organic Light-Emitting Diodes (OLEDs) with Doping Iodine and Nitrogen Treatment

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Abstract-Simultaneously doping iodine (I_2) inorganic dopants and nitrogen (N_2) treatment on hole transport layer (HTL) to promote the output luminance of the Al/ Tris-(8-hydroxyquinoline) aluminum (Alq_3)/N, N'-diphenyl-N, N'bis (3-methylphenyl)-1, l'-bipheny-4, 4'-diamine (TPD)/ITO organic light-emitting diodes (OLEDs) has been studied in detail. Experimental results show that the output luminance can be significantly promoted up to 950% and 560% in magnitude at bias of 8V and 10V, respectively. We attribute the obvious promotion to significant morphological changes occurring in HTL as a result of the di-nitrogen occlusion and I_2 dopant generated guest hopping sites. The morphological changes improve the HTL/Alq3 interface while the interfering action between guest and host hopping sites lowers hole mobility thus raising the output luminance in the OLEDs.

KEYWORDS: organic light-emitting diodes (OLEDs), morphological, guest hopping sites, interfering action

1.Introduction

With the features of low power dissipation, wider vision angle, high luminescence, shorter response time and simplified fabrication, organic light electro- luminescence diodes (OLED) have been paid much attention for flat panel display[1]. In the past, use of multiple-layer structures or multiple-quantum-well (MQW)[2-3], addition of both organic materials as well as phosphor-sensitized fluorescence[4] and employ of the assisted dopant[5] have been proposed to enhance the output luminance of OLED. On the other hand, we have tried doping iodine in HTL layer to create guest-hopping sites, which then interfere the hopping transport of holes via the host sites thus achieving the effective controlling the number of hole for the recombination. In this letter, we report the study in detail. Experimental results showed the luminance of a bi-layer OLED with the structure Al/Alq_3(Tris-(8-hydroxyquinoline) Aluminum)/TPD(N, N'-diphenyl-N, N'bis (3-methylphenyl)-1, l'-bipheny-4, 4'-diamine)/ITO was dramatically promoted by doping iodine in TPD. Furthermore, the luminance could be raised more if the doping is under N2 ambient, for example, the promotion in output luminance could attain to 950% and 560% in magnitude at bias of 8V and 10V, respectively. We attribute the obvious promotion to the significant morphological improvement in TPD/Alq3 interface as a result of the di-nitrogen occlusion [6]. In comparison to the reported technologies, the developed method possesses the advantages of low cost and popular doping materials and needs no any extra process in preparation of device.

2. Device Fabrications and Experimental

The bi-layer OLEDs samples with area of 0.7cm x 0.7cm and the configuration of Al/Alq_3/TPD /ITO were prepared in the glass substrates pre-coated by indium tin oxide with sheet resistance $R_s \leqq 10\Omega$/square .Firstly evaporate 500Å TPD on cleaned ITO as HTL under $1x10^{-5}$ Torr, Various HTLs such as TPD only, iodine doped TPD with and without N2 ambient($1x10^{-4}$ Tor) were employed. The iodine doped TPD layers were prepared in various weight ratios (i.e., I_2/TPD =5/1, 10/1 and 15/1) Next, sequentially evaporated 500 Å Alq_3 and 1500Å Al as ETL and cathode, respectively. Finally, HP4156 and TOPCON BMP were used to measure the electrical characteristics, output luminance, respectively.

3.Result and Discussion

Figure 1 shows the Luminance-Voltage (L/V) curves of the OLED with various TPD evaporation conditions, while the inset gives the zoom of the curves under lower bias. As seen, the output luminance increases with increase of I_2/TPD weight ratio up to 10/1 (curve c) then decreases for 15/1 weight ratio (curve d). It is the doping of I_2 generates the guest hopping sites in TPD layer[7-8], thus interfering the hoping transport of holes and in turn the OLEDs' output luminance. The detailed mechanisms are illustrated schematically in Fig.2. In which, (a) presents the holes hopping in TPD without doping, and (b) to (d) for holes transport via both guest sites and host sites (HOMO of TPD) alternatively under conditions of lightly concentration doping, optimum concentration doping and heavy concentration doping, respectively. In the case of lightly doping of I_2 (Fig. 2b), the holes transport not only via both gust hopping sites and host hopping sites but it also jump between the gust hopping sites and host hopping sites (called interfering action). However, most of the holes are still hopping via the host hopping sites and interfering action is not significant, because of the number of guest hopping site is quiet smaller compared to that of the host sites The small number of guest hopping site results in larger distance between the guest hopping sites thus retarding the hole hopping. Until the doping quantity is up to an optimum value (in this case, 10/1 for I_2/TPD), as shown in Fig.2(c), the interfering action became obvious due to the shrank distance between guest hopping sites is comparable to that of the host sites now. The interfering action interrupts the holes transport significantly and in turn enhancing the balance of electron hole pair to recombine in the interface. However, as the dopant concentration over the optimum value (Fig. 2(d)), the more shrank distance between guest site speeds the holes transport in TPD thus lowering the output luminance. Additionally, as shown in Fig.3 and its inset, a significantly lowering of roughness from 1.434 nm to 0.576nm can be found in base of the atomic force microscopy (AFM) analyses for the TPD films' morphology with and without nitrogen under N_2 $1x10^{-4}$ Torr ambient. The smooth TPD improves the contact of Alq_3/TPD layers and in turn reducing the generation of traps and heat in interface, which has been reported as the key killers of OLEDs' output luminance [9]. The significant morphological improvement is attributed to the di-nitrogen occlusion in TPD layer[6]. Both iodine doping and N_2 treatment promote OLEDs' output luminance up to 950% and 560%, under bias at 8V and 10V, respectively as shown in curve (e) of Fig. 1.

The interfering action between the gust hopping sites and host hopping sites model, the current density is nicely supported by the current density v.s. voltage curves (J/V curve) in linear and log-log scale for split samples are shown in Fig. 4 and its inset, respectively. As seen, firstly the current density increases with bias to the maximum at voltage Vp(around 4.0 V) due to the bias enhanced injection of carriers for all samples. But the improving output luminance is not obviously as shown in the insert of Fig.1 due to the injected holes and field are still small for interfering action, which need energy to assist holes jump from HOMO level into gust hoping sites. As bias over the peak voltage Vp, holes in HOMO level now get enough energy to jump and hop alternatively between the guest level and HOMO level, i.e., starting the interfering action thus lowering the current and generating a negative differential resistance (NDR) in the J/V curve. In which, Vp_{I_2}, $Vp_{I_2+N_2}$ and Vv are the peak and valley voltages of NDR for sample with and without N_2. On the other hand, based on the log-log scale in the inset, we found for undoped samples, following the increase of bias, the curve starts with space charge limited (SCL) then becomes the trap charge limited (TCL) dominated and finally it goes back to SCL dominated, this is same as that has been reported previously [10]. In iodine-doped samples, the J-V curves start with SCL then turn to TCL dominated as the undoped one. However due to the generation of guest hopping sites, the NDR is found in J-V curve and the curve doesn't back to SCL dominated again. It is worthy to note, in comparison both Fig.1and Fig. 4, the output luminance becomes significantly in the bias of 4.0V which also the starting voltage of NDR. The well corresponding between Figs. 4 and 1 provides another vigorous supported more to the model of Fig. 2., i.e., the interfering action can effectively degrade the holes transport thus lowering the current and raising output luminance due to more balanced carrier's number for recombination. Beyond the Vv (the valley voltage of NDR), both raise in current and output luminance are supposed to the injection of carriers is larger enough to suppress the effect of interfering action at this time. In summary, the NDR is a sign of significant interfering action of holes well corresponding between Figs. 4 and 1 provides another vigorous supported more to the model of Fig. 2.

4.Conclusion

In summary, co-evaporation of HTL with I_2 under N_2 ambient to promote the luminance of OLED has been studied in detail. Experimental results show the promotion of output luminance up to 950% and 560%, under bias at 8V and 10V, respectively .The promotion is come from the successful manipulating the hole hopping transport in guest and host hopping site thus enhancing the balance of electron-hole pairs to recombine in the interface of Alq3/TPD. Simultaneously the di-nitrogen occlusion in TPD layer improves the contact of Alq_3/TPD layers thus reducing the generation of traps and heat in the interface. Compared to the reported technologies, the developed inorganic molecular technology possesses the advantages of low cost and addition no any extra process in preparation of device. Therefore, the developed method is a potential technology to raise the luminance of OLED.

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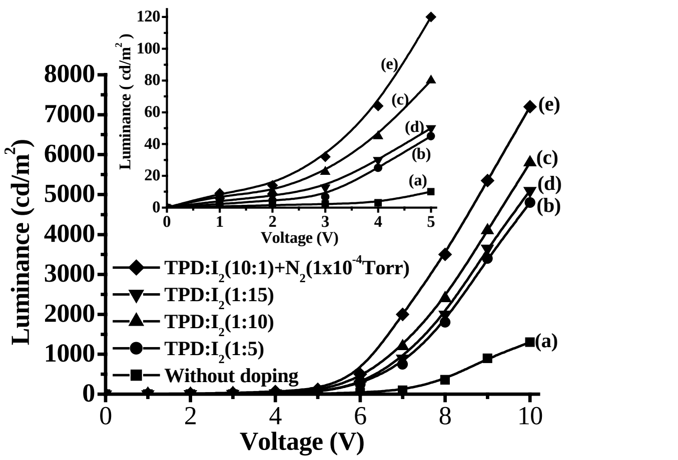
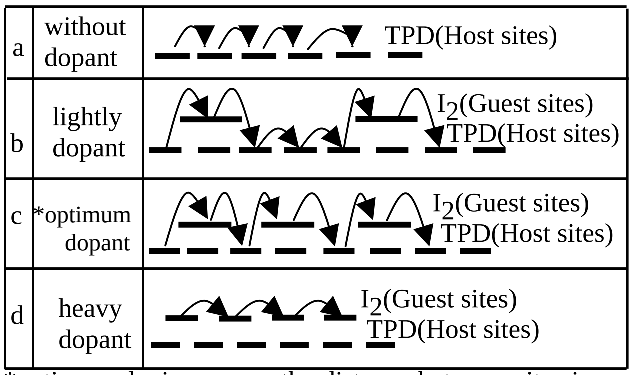


Fig.1.The output luminance of OLEDs as a function of driving voltage for split HTL evaporation conditions; curve (a) without doping, curve (b) with weight ratio TPD :I_2 =1:5, curve (c) with weight ratio TPD :I_2 =1:10, curve (d) with weight ratio TPD :I_2 =1:15 and curve (e) with weight ratio TPD :I_2 =1:10 under N_2 $1x10^{-4}$ Torr ambient. The inset shows the zoom for the driving voltage 0 to 5V



*optimum doping means the distance between sites is optimum to obtain the maximum interfering action

Fig.2. The models for holes transport in the host sites and guest sites with iodine dopant; (a) without doping, (b) with lightly iodine dopant, (c) with optimum iodine dopant, (d) with heavy iodine dopant

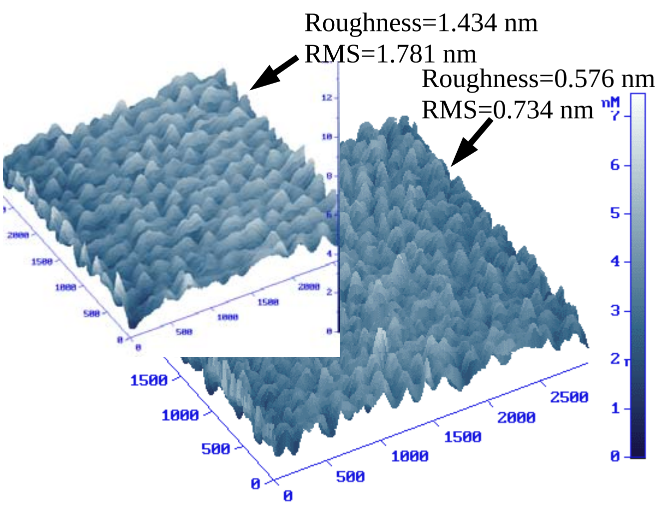


Fig.3. AFM micrographs of iodine doped TPD film with $1x10^{-4}$ Torr nitrogen ambient and without (insert)

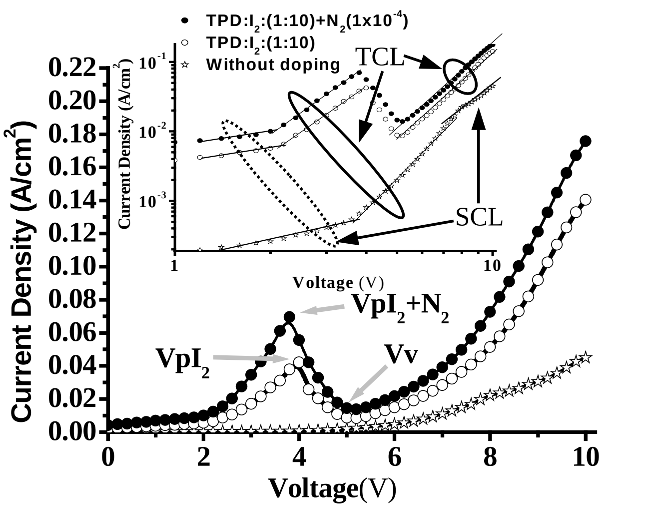


Fig.4. The current density-voltage curves with linear and log-log scale (inset) of OLEDs with iodine dopnat under nitrogen ambient (TPD/ I_2 =1/10 and N_2 $1x10^{-4}$ Torr ambient), the OLEDs with iodine dopnat (TPD/ I_2 =1/10) and without iodine dopant are included.