Fabrication of flexible organic light-emitting diodes with Alq_3 as emitting layer*

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Fabrication of flexible organic light-emitting diodes(FOLEDs) with ITO/PVK:TPD/Alq_3/Al configuration prepared on PET substrates is reported. Alq_3 is used as the light-emitting material. The curves of the current density vs. voltage, optical current vs. voltage and quantum efficiency vs. current density of the devices are investigated. Compared the devices with the ones that have the same configuration and are fabricated under the same conditions but on glass substrates, the characteristics of the two kinds of devices are very similar except that the threshold voltage of the flexible FOLEDs is a little higher. Under the driving voltage of 20V, the corresponding brightness and the external quantum efficiency are 1000 cd/m^2 and 0.27%, respectively. In addition, the anti-bend ability of the devices is tested and the reasons of failure of the devices are analyzed.

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Organic light-emitting diodes (OLEDs) are promising devices in the field of information display because they can offer low power consumption, wide viewing angle, good contrast, and video rate operation[1,2]. Since the ITO film in most of the presente OLEDs are fabricated on solid glass substrates, these OLEDs have the disadvantages of inflexibility, brittleness and heaviness. Actually these disadvantages can be overcame by simply fabricating the OLEDs on various kinds of substrates including polymers, i. e. a FOLED[3-5]. The flexible substrates will significantly reduce the weight of OLEDs and provide the capabilities of conform, bend or rolling a display into any shapes. Moreover, it will open the possibility of fabricating displays by continuous roll processing, thus providing the basis for cost-effective mass production. In this paper the fabrication of FOLEDs with Alq_3 as the emitting layer and flexible ITO film as anode is reported. The measurement results of the devices are compared with the ones that prepared under the same conditions but with ITO film on glass substrates. Especially the anti-bend ability of the devices is tested and analyzed.

Two kinds of devices are fabricated on PET (devices I) and glass (devices II) substrates coated with ITO using a conventional vacuum vapor deposition and spin-coating. The configurations of the two kinds of devices are respectively as the follows:

I: PET/ITO/PVK:TPD/Alq_3/Al

II: Glass /ITO/PVK:TPD/Alq_3/Al

where PVK:TPD is used as the hole transport layer (HTL), Alq_3 as the emitting and electron transport layer, and aluminum (Al) as the cathode. The molecular structures of the organic material and the configuration of the device are shown in Fig. 1.

For ITO film on PET, the average optical transmittance (in the wavelength range of 400 nm~800 nm) is more than 80% and the sheet resistance is less than 170 Ω/□. The total thickness of PET substrate is about 125 μm. The process of preparing both devices is the same. Firstly, ITO film is etched to stripe and clean. Secondly, the TPD blended with PVK in chloroform solvent is spin coated onto ITO-coated substrate at 1 000 rpm for 18 s and then at 4 000 rpm for 30 s. The weight ratio between PVK and TPD is 1:1, and the total concentration is 1.5 mg/ml. Thirdly, Alq3 is deposited by means of vacuum vapor deposition in a vacuum of 7×10^{-4} Pa. Finally, Al is evaporated as a cathode through a shadow mask.

In addition, the EL spectra and brightness are all measured with Photo Research PR650. The characteristic curves of current density vs. voltage and quantum efficiency vs. current density of the devices are measured by the system with Keithley 2400 and Keithley 485[6]. The active emissive area of the device is 2 mm×2 mm. And all the measurements were carried out with unpackaged devices at room temperature.

The reason for using TPD-blended PVK is that the polymeric compound PVK can prevent the low molecular mass material TPD from being degraded by aggrega-

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tion. Besides, PVK can also smooth the ITO surface and improve the ITO surface morphology obviously, which will enhance the capability of hole-injection. On the other hand, the classic hole transport material TPD not only can effectively reduce the interfacial barrier height between the organic layer and the anode, but also can prevent the electrons from sliding away from the emitting layer. As a result, the stability and lifetime of the FOLEDs can be improved obviously[7,8]. The reason for omitting the electron transport layer in the design of FOLED configuration is that the emitting material Alq_3 has been proved to have a better electron transporting property[2]. Therefore, the fabrication processes of FOLEDs are greatly simplified.

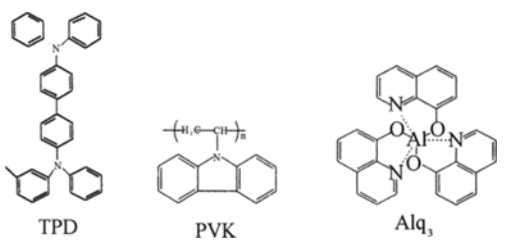

Fig. 1(a) Molecular structures of TPD, PVK and Alq_3

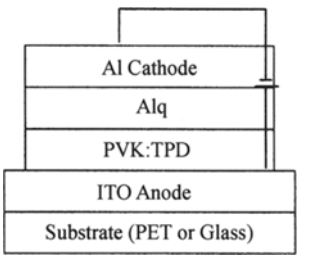

Fig. 1(b) The device configuration

Fig. 2 shows the EL spectrum of both devices, and the main peak is located at 540 nm. The EL spectrum of the devices is the same as the characteristic spectrum of Alq_3. It proves that the emitting of both devices comes from Alq_3.

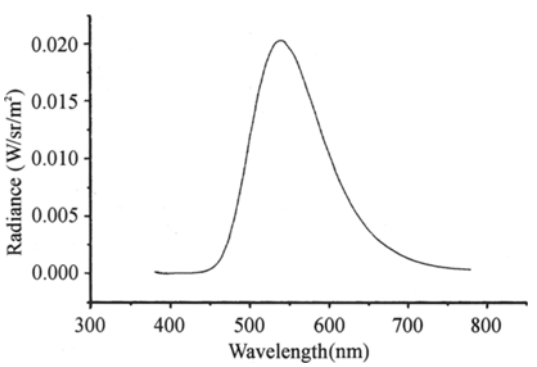

Fig. 2 The EL spectrum of Alq_3

Fig. 3 shows the typical current density versus voltage (J-V) characteristic curves for both devices. As shown in Fig. 3, it can be found that the current density power output is proportional to the exponential of driving voltage. The J-V curves show a typical diode behavior of both devices. It is found from the J-V curves that the threshold voltage is almost the same (about 6-7 V) for both devices, but the current density of the devices I increases more slowly than that of devices II with increasing of the bias.

Fig. 4 shows the optical current versus voltage characteristic curves of both devices. For the devices I, an optical current of 2.83×10^{-10} A can be obtained at an applied voltage of 7 V, and under this circumstance the light emitting of the devices can be observed by naked

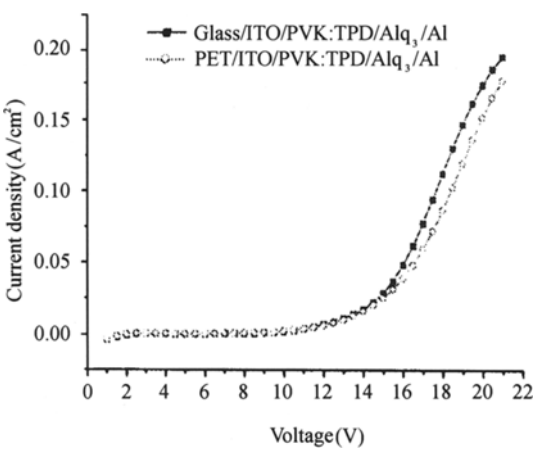

Fig. 3 $J-V$ curves of devices I and II

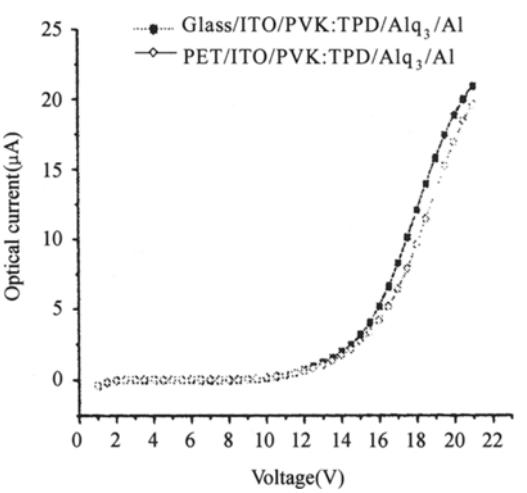

Fig. 4 Optical current vs. voltage of devices I and II

eye, while the same optical current is obtained at voltage of 6 V for devices II. The threshold voltage of devices I is about 7 V, which is a little higher than that of devices II (6 V). A brightness level of 1000 cd/m^2 is obtained at 20 V for the devices I, while 1 200 cd/m^2 is observed for devices II. Both devices emit bright green light. When the external bias is not much larger than the threshold voltage, light emitting of the two kinds of the devices is homogeneous and stable. But with farther increasing of the bias, the brightness of the devices II increases more obviously than that of the devices I.

The evolution of both devices external quantum efficiency at different applied current densities is shown in Fig. 5. We can conclude from the curves that the emission efficiency of both devices increases rapidly at first, and then slowly flats out. The maximum external quantum efficiency of devices I is approximately 0.27% at about 0.4 A/cm^2, for which the driving voltage is 20 V, while that of devices II is approximately 0.29 % at about 0.4A/cm^2.

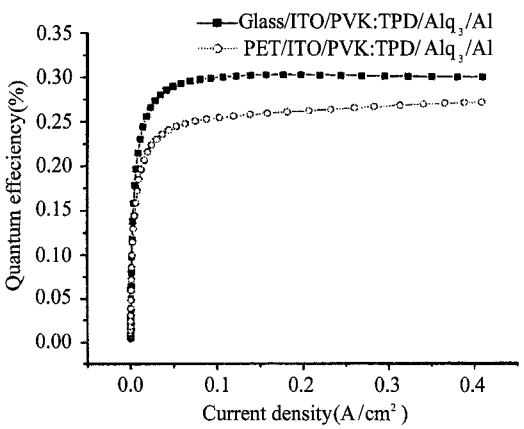

Fig. 5 ***QE-J*** **curves of devices I and II**

It can be seen that the curves of *J-V*, optical current vs. voltage and *QE-J* of the devices I are not obviously different from those of the devices II. But the performance of devices I is slightly poorer than devices II. For devices I, when the applied voltage is increased over a certain value, although the samples still emitting green light, it is found that bursting occurs from many distributive dots on the surface of devices I. If the bias increases further, the number of flare-up dots increases and the dots diffuse over the surface. Correspondingly the current through the sample decreases rapidly and the devices fail. For the devices II, however, no flare-up phenomenon is observed, even when the bias is increased up to 25 V. Although there are some black spots on the surface with the increasing working time, it doesn t mean the failure of the device. Because if the external bias is switched off and turned on again, the sample could still work. Of course, when the bias is high enough or the working time is long enough, the devices II will fail at last. We find that the black spots are constantly enlarged until eventually lead to failure[9,10].

The main reasons that cause the performances of PET devices come down are as the followings. The fabricated condition of ITO films impacts on the crystal structure, surface properties, photo-electrical properties and probably work function of ITO films, and hence influences the performances of the devices. It is found that the main differences of the ITO film on PET from that on glass are the adhesion and surface roughness. The capability of ITO film adhering to PET is less than that to the glass. The less adhered ITO film is easy to be peeled from the substrates when the device is working in high voltage, and thereby brings out the bursting of the PET devices. The surface roughness of ITO film on PET is larger than that of ITO film on glass. The apparent larger concave in the surface may come from defects of the substrate. Such surface roughness induces poorer film quality of the PET devices than that of glass. When the devices work, a higher electric field intensity due to smaller organic thickness readily induces the damage of the organic layer[9,10]. In addition, some black spots are resulted from the delamination of the metal at the Alq_3/Al interface and initiated by pinholes on the cathode. The pinholes are formed by defects introduced during device fabrication[9]. Moreover, it can be seen that there are many obvious nicks in the PET devices. These nicks probably arose from the PET substrate scathed in the clean process, which obviously play a negative role in the uniformity of the organic films.

Furthermore, the anti-bending ability of PET devices curved are tested and analyzed. The devices worked under bias 15 V during the experiment. The data are shown in Table. 1. As shown in it, when devices I have been bended by 80 times, the brightness comes down less than 50% of that it was. When the devices have been bended more than 100 times, the devices failed. When the devices have been bended repeatedly, many nicks arise on the anode which can be observed by naked eyes, and delamination phenomena of the metal appeares at the Alq_3/Al interface. All those mentioned above would be the reasons that lead to the damage of the devices[9,10].

Tab. 1 Experimental data(bias: 15 V)

Times of bending	Brightness (cd/m^2)	Ratio to original Brightness (%)
0	600	100
20	551	91.8
40	462	77.0
60	362	60.3
80	251	41.8
100	180	30.0

FOLEDs with ITO/PVK: TPD/Alq_3/Al configuration prepared on PET substrates have been fabricated. The curves of the current density vs. voltage, optical

current vs. voltage and quantum efficiency vs. current density of the devices are investigated.

Compared with those that have the same configuration and are fabricated under the same conditions but on glass substrates, the characteristics of the two kinds of devices are very similar except that the threshold voltage of the FOLEDs is a little higher. Under the driving voltage of 20 V, the corresponding brightness and external quantum efficiency of FOLEDs are 1 000 cd/m^2 and 0.27 %, respectively. In addition, the mechanism of luminescence and the reason of failure of the devices are discussed. The anti-bending ability of the devices are tested and analyzed. Fig. 6 shows a picture of FOLED when it was working and kept bended. The picture was taken in a dark room.

References

[1] Friend R. H., Gymer R. W., and Holmes A. B. *Nature*, **397** (1999): 121.

[2] Tang C. W., and Vanslyke S. A. *Appl. Phys. Lett.*, **51** (1987): 913.

[3] Sirringhaus H., Tessler N., and Friend R. H. *Science*, **280** (1998): 1741.

[4] Gustufsson G., Cao Y., and Treacy G. M. *Nature*, **357** (1992): 477.

[5] Gu G., Burrows P. E., and venkatesh S. *Opt. Lett.*, **22** (1997): 172.

[7] Hua Yulin and Li Juan. *J. of Optoelectronics • Laser*, **15** (2004)516. (in Chinese)

[6] Zheng Jiajin, Hua Yulin, and Yin Shougen. *Chin J Lumin*. **25** (2004)231. (in Chinese)

[8] Young K. W., Chung C. H., and Lee J. H. *Thin Solid Films*, **426**(2003)246.

[9] Zhao J. Q., Xie S. J., and han S. G. *Physica Status Solidi (A)*, **184**(2001)233.

[10] Wang shuguo, Hua Yulin, and Zheng jiajin. *J. of Optoelectronics • Laser*, **15**(2004)663. (in Chinese)