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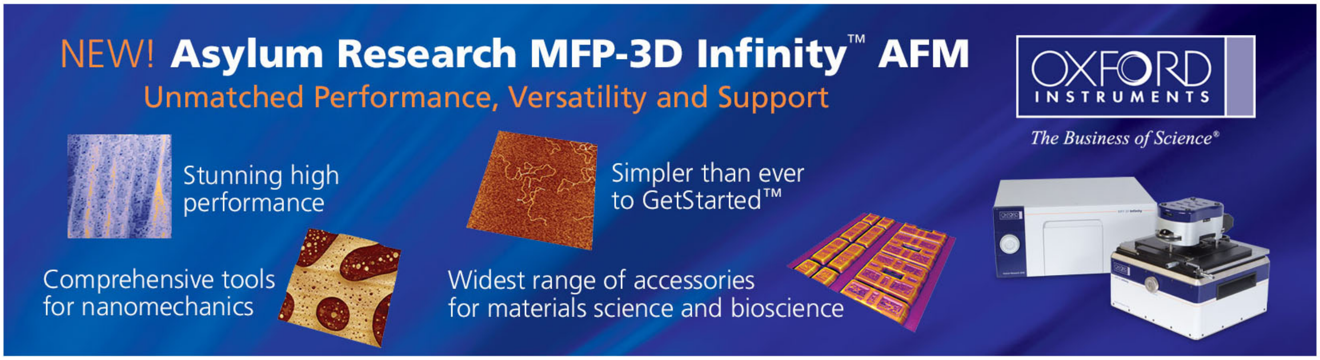
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Highly efficient blue organic light-emitting devices incorporating a composite hole transport layer

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Organic light-emitting devices (OLEDs) incorporating the copper phthalocyanine (CuPc)/ *N*,*N*′-*bis*-(1-naphthyl)-*N*,*N*′-diphenyl-1,1′-biphenyl-4,4′-diamine composite hole transport layer have been fabricated. Under optimal condition, the highly efficient sky-blue OLED based on the aminosubstituted distyrylarylene fluorescent dye, doped in the stable diphenyanthracene blue host achieved a luminance efficiency of 16.2 cd/A with a Commission Internationale d' Eclairage $(\mathrm{CIE})_{x,y}$ color coordinate of [0.15, 0.29] at 6.4 V ($20\ \mathrm{mA/cm^2}$) and an external quantum efficiency of 8.7%. © *2005 American Institute of Physics*. [DOI: 10.1063/1.1931052]

Since Tang and VanSlyke developed multilayer organic light-emitting devices (OLEDs),[1] tremendous efforts have been directed toward improving the device performance. It was recognized that the external quantum efficiency (EQE) of OLEDs depends heavily on the efficiency of carrier injection and recombination as well as the balance of the holes and electrons.[2,3] Therefore, in order to achieve maximal efficiency, a balanced carrier recombination in the emission layer is a must in the device. OLEDs in general do not necessarily provide the configuration to achieve a balanced carrier injection/transportation that leads to recombination. One of the reasons is that the injected hole is usually more mobile than the injected electron under the same electric field, therefore, finding a way of reducing the number of holes or increasing the number of electrons reaching the emission layer is considered one of the most direct and economic solutions to improve device efficiency.

Up until now, several approaches including modification of the anode/cathode contact or hole/electron transport layer have been known to improve carrier balance and recombination.[4,5] One of the most simple and often used methods is to interpose a buffer layer, such as copper phthalocyanine (CuPc), between an indium tin oxide (ITO) anode and a hole transport layer (HTL),[6] which can improve the device efficiency and stability. However, using CuPc as a buffer layer can lead to substantial increase in undesirable device operation voltage. The increase in device efficiency induced by inserting a CuPc buffer layer is correlated with a decrease in hole-injection efficiency, which leads to a better recombination of holes and electrons.[7] Besides inserting a buffer layer in-between the anode and HTL, it has been reported that doping of "hole-trapping" materials, such as rubrene, in HTL like *N*,*N*′-*bis*-(1-naphthyl)-*N*,*N*′-diphenyl-1,1′-biphenyl-4,4′-diamine (NPB) also brought about similar effect.[8] However, the increase in device efficiency as well as device operation voltage is not as significant. This can be attributed to the different nature of the hole-impeding process in both cases. The CuPc buffer layer impedes the injection of holes into the HTL, but the usual hole-trapping dopant molecules tend to reduce their mobility inside the HTL and as a result, the effect of which may not be as strongly reflected on the current density–voltage (*J*-*V*) characteristics because of the superior hole mobility characteristics of NPB.[9]

Recently, we have combined the advantage of CuPc with the doping technique and developed an OLED structure having a CuPc/NPB composite HTL (*c*-HTL) (Ref. 10) which could simultaneously maintain the advantages of the hole-impeding property of CuPc as well as the low voltage characteristic of a doped device. In this letter, we will demonstrate that this OLED structure can significantly enhance the device efficiency by controlling the hole mobility without compromising much of its device operation voltage. The sky-blue OLED shown in this letter has also delivered a luminance efficiency of 16.2 cd/A with a Commission Internationale d'Eclairage $(\mathrm{CIE})_{x,y}$ color coordinate of [0.15, 0.29] at 6.4 V ($20\ \mathrm{mA/cm^2}$) and an EQE of 8.7%.

In our experiments, two blue devices have been fabricated. Device I is a standard blue device without *c*-HTL, whereas Device II is the blue device incorporating *c*-HTL. The detail molecular structure and device architecture is shown in Fig. 1, where CF_x is the hole-injection layer,[11] *c*-HTL and NPB as a whole is the HTL, *p*-*bis*(*p*-*N*,*N*-diphenyl-aminostyryl)benzene-doped 2-methyl-9,10-*di*(2-naphthyl)anthracene (MADN) is the blue emitter, *tris*(8-quinolinolato) aluminum ($\mathrm{Alq_3}$) is the electron transport layer, and LiF/Al is the bilayer cathode. After the routine cleaning procedure of ultrasonicating the ITO coated glass in organic solvents, deionized water, and dried in oven at 120 °C, the ITO substrate was then loaded on the grounded electrode of a parallel-plate plasma reactor, pretreated by oxygen plasma, and then coated with a polymerized fluorocarbon film. Devices were fabricated under the base vacuum of about 10^{-6} Torr in a thin-film evaporation coater following a published protocol.[12] A multilayer structure of *c*-HTL/NPB/emissive layer (EML)/$\mathrm{Alq_3}$/LiF/Al was deposited on the substrate by resistive heating with a thickness

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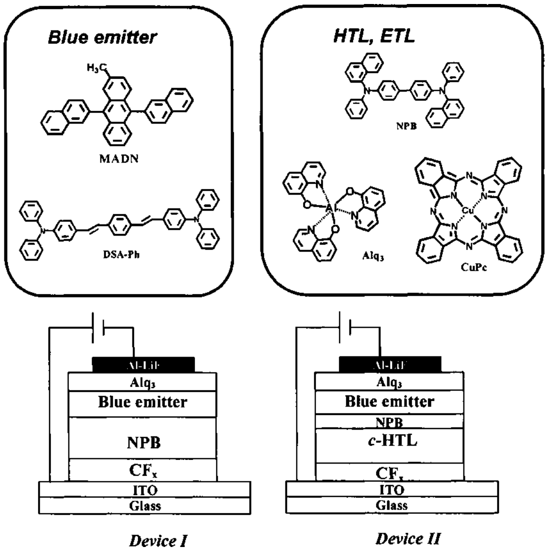

FIG. 1. Molecular and device structures used in the experiment.

of 40, 30, 40, 10, 1, and 200 nm for *c*-HTL, NPB, EML, Alq_3, LiF, and Al, respectively. The purpose of inserting a layer of pure NPB in-between *c* -HTL and EML is to block electrons from reaching *c*-HTL since the lowest unoccupied molecular orbital of CuPc (~3.0 eV) is lower than that of MADN (2.5 eV). It will give rise to nonradiative recombination in CuPc, which has been proven to produce low luminance efficiency of the device.[10] In the evaporation of *c*-HTL, CuPc and NPB were codeposited at the desired ratio (1:1), which was determined previously.[10] All devices were hermetically sealed prior to testing. The active area of the electroluminescence (EL) device, defined by the overlap of the ITO and the cathode electrodes, was 9 mm^2. The current-voltage-luminance characteristics of the devices were measured with a diode array rapid scan system using a Photo Research PR650 spectrophotometer and a computer-controlled programmable dc source. Besides the two blue devices, three hole-only devices have also been fabricated to investigate the hole-transporting property of different CuPc:NPB compositions (0:100, 12:88, and 50:50). The structure and energy level diagram of hole-only devices is shown in the inset of Fig. 3, where the low-work function Au (~5.3 eV) was used as the cathode to prevent electrons from injecting into the organics.[13] As a result, holes are the dominant, if not the only carriers in the organic layer when a voltage is applied across the device.

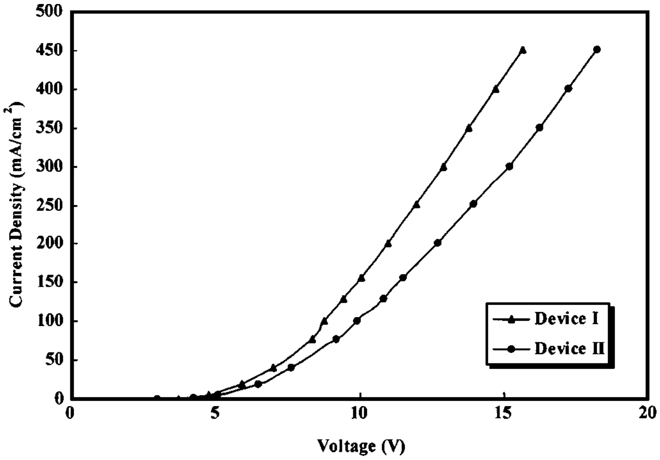

FIG. 2. *J*-*V* characteristics of the blue devices.

Figure 2 shows the dependence of *J*-*V* characteristics of the blue devices. Higher operation voltage was observed in the device with *c*-HTL comparing to that of the standard blue device. For instance, the operation voltage at 20 mA/cm^2 is 6.4 V for *c*-HTL device and 5.9 V for the standard blue device. We believe that by incorporating *c*-HTL, the hole-transporting property was reduced. More direct evidence of reduced hole mobility by *c*-HTL can be observed from the *J*-*V* characteristics of hole-only devices shown in Fig. 3.

It is noted from Fig. 3 that the voltage required at a given current density of *c*-HTL, the hole-only device is higher than that of the pure NPB device, which in other words, means the transport of holes in *c*-HTL is more difficult compared to that in the pure NPB. It is also noteworthy that the voltage was lowered when a high doping level of

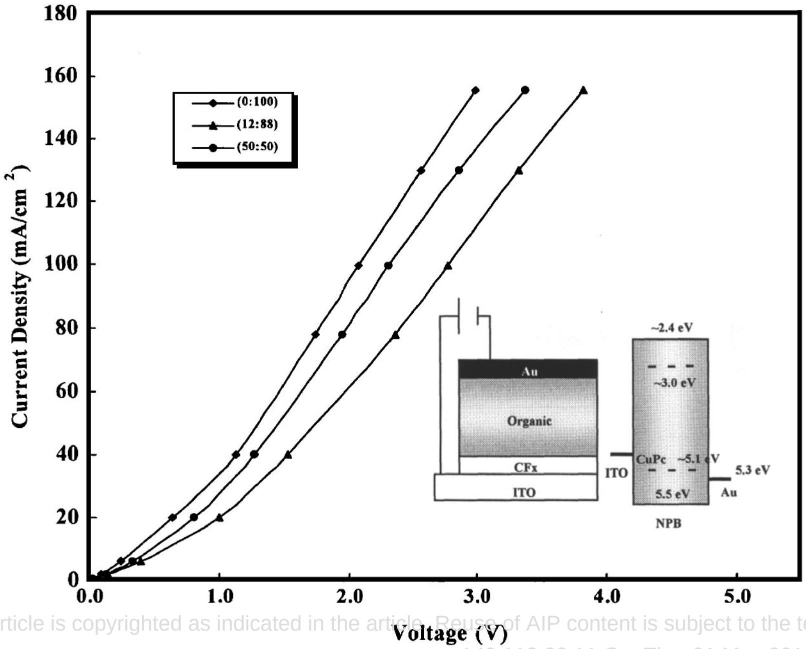

FIG. 3. *J*-*V* characteristics of hole-only devices (inset is the hole-only device structure and energy level diagram).

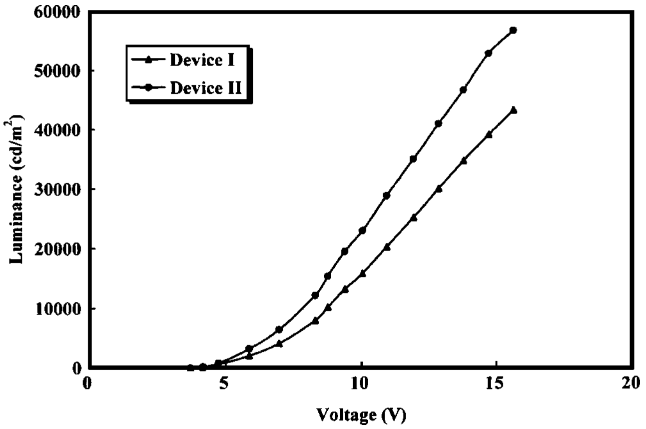

FIG. 4. *B-V* characteristics of the blue devices.

CuPc was used (50% CuPc). This phenomenon may be rationalized by the following explanations. We believe that the increase in operation voltage upon incorporating *c*-HTL is due to two contributing factors. One is the "hole-impeding" character of CuPc, which slows down the hole transport, hence, increases the operation voltage. The other contribution is the dissipation of energy from the "trapping and detrapping" mechanism of holes on the highest occupied molecular orbital of CuPc and NPB since the ionization potential of CuPc (~5.0 eV) is much lower than that of NPB (~5.4 eV). By increasing the concentration of CuPc in the *c*-HTL, the propensity for transport by undergoing trapping and detrapping will be much reduced due to the shorter intermolecular distances amongst CuPc molecules.[14] As the result, holes can hop between the CuPc molecules, and hence lower operation voltage. Besides these two possible routes, holes can also hop between the NPB molecules, which would impact little on the operating voltage due to the superior hole mobility characteristics of NPB.[9] Therefore, by adjusting CuPc doping level, the hole mobility and device operation voltage could be optimized simultaneously.

Figure 4 shows the dependence of luminance-voltage (*B-V*) characteristics of the blue devices. Considerably higher luminance was observed in the device with *c*-HTL compared to that of the standard blue device, which we believe is due to the reduced hole mobility by *c*-HTL that results in more balanced charge carriers and carrier recombination at the emitting zone. The luminance at 20 mA/cm^2 is 3229 cd/m^2 for the *c*-HTL device and 2074 cd/m^2 for the standard blue device.

The overall EL performances of these blue devices are summarized in Table I. Compared with the luminance efficiency of the standard blue device without *c*-HTL (10.4 cd/A), that of the device with *c*-HTL has been improved 60% to 16.2 cd/A with a CIE coordinates of (x=0.15, y=0.29) and a power efficiency of 7.9 lm/W (20 mA/cm^2) at 6.4 V. The incorporation of *c*-HTL not only produces a substantial increase in device efficiency, but also emits color that is essentially identical to that of the device without *c*-HTL.

TABLE I. EL performances of blue devices driven at 20 mA/cm^2.

Device	Luminance (cd/m^2)	Voltage (V)	1931 CIE X	1931 CIE Y	Lum. Yield (cd/A)	Efficiency (lm/W)	EQE (%)
I	2074	5.9	0.15	0.30	10.4	5.6	5.3
II	3229	6.4	0.15	0.29	16.2	7.9	8.7

Carrier recombination, as well as the balance of holes and electrons, is considered to be one of the most important factors that determine the EQE of OLEDs. Here, we have developed a simple method of introduction of the *c*-HTL, which can efficiently control the hole mobility in order to improve hole-electron balance. Improvement in the hole/electron recombination efficiency was illustrated by the high EQE and luminance efficiency of 8.7% and 16.2 cd/A, respectively.

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