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Improved efficiency for green and red emitting electroluminescent devices using the same cohost composed of 9,10-di(2-naphthyl) anthracene and tris-(8-hydroxyquinolinato) aluminum

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ABSTRACT

We demonstrate highly efficient green and red fluorescence dyes-doped electroluminescent devices using cohost strategy. The cohost system is composed of tris-(8-hydroxyquinolinato) aluminum (Alq) and 9,10-di(2-naphthyl) anthracene (ADN). The maximum current efficiencies are increased by 54% and 104% for green and red devices by optimizing the ratio between ADN and Alq in the cohost compared to the conventional Alq single-host devices, respectively. We attribute the improvement of efficiencies to balanced hole and electron injection into the emitting layer, the enlarged width of recombination region and the multiple emission processes.

1. Introduction

Organic light emitting device (OLED) is a promising technology for next-generation displays and solid-state lightings due to its simple fabrication process and flat-emission capability. One of the key contributions in the development of OLED technology can be attributed to the discovery of the guest–host-doped emitter system [1]. This is because a single-host material may be used together with a variety of dopants leading to electroluminescence (EL) of desirable hues with high efficiencies and extended operational stability [2]. However, it is found that some guest–host systems cannot achieve the expected results. Therefore, the assistant dopants have been introduced into the guest–host systems to improve the EL performances of the OLEDs [3,4]. But there also exist some disadvantages that cannot be removed. In OLEDs, the host materials should have the abilities of transferring energy efficiently to the dopants and the bipolar transporting character. If the energy transfer ability is poor, the energy cannot be used by the dopants completely, resulting in the emission of the host and the low efficiency of the device. And the poor bipolar transporting ability could lead to the narrow recombination region, which is a primary cause for EL emission loss and shortening of the operating lifetime of the devices [5]. However, host materials that could simultaneously fulfill the two requirements are rather rare. Thereby, the OLEDs using a single host usually cannot result in very high performances. Thus, to overcome the disadvantages, the cohost structure has been used for the construction of OLEDs [6,7]. Liu et al. fabricated 4-(dicyanomethylene)-2-*t*-*butyl*-6-(1,1,7,7-tetramethyljulolidyl-9-enyl)-4*H*-pyran (DCJTB)-doped OLEDs, in which the blend of 5,6,11,12-tetraphenylnathacene (rubrene) and tris-(8-hydroxyquinolinato) aluminum (Alq) was used as the cohost. They attributed the improved EL efficiency and lifetime to the bipolar transporting character of rubrene in the cohost system [6]. Lee et al. studied the devices using the mixture of bis(10-hydroxybenzo[h]qinolinato) beryllium and N,N′-diphenyl-N,N′-bis(1-naphthyl)-(1,1'-biphenyl)-4,4'-diamine (NPB) as the cohost and 9-benzothiazol-2-yl-l,l,6,6-tetramethyl-2,3,5,6-tetrahydro-1H,4H-11-oxa-3a-aza-benzo anthracene-10-one (C-545T) as the dopant. They also ascribed reduced the driving voltage and the increased lifetime to the bipolar transporting character of the cohost [7]. Though the performances of the OLEDs have been improved, each previously reported cohost structure focused on only one dopant. So the practicalities of the cohost systems are

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largely restricted. If a cohost structure could be doped with various monocolor emission dyes, we would provide much simpler fabrication processes of the full-color displays and illuminations based on OLEDs.

In this paper, we fabricate highly efficient green and red fluorescence OLEDs using the same cohost system. When designing the cohost system, we compare the carrier transporting properties, the lowest unoccupied molecular orbital (LUMO) and the highest occupied molecular orbital (HOMO) levels among several general host materials. It is found that 9,10-di(2-naphthyl) anthracene (ADN) is a bipolar transporting host but could only be used as the suitable host for blue emission dopants because of its wide band-gap. When it was used as the host for green or red emission dopants, the ADN emission would appear in the EL spectrum for the inefficient energy transfer from ADN to the dopants, especially the situation at the higher driving voltages. This would largely affect the color purity of the light. Alq exhibits a lower band-gap but unipolar carrier transporting character relative to ADN. Thus, the two traditional fluorescence hosts, ADN and Alq, are used as the two components in the cohost system. By optimizing the ratio between ADN and Alq in the cohost, the maximum current efficiencies are increased by 54% and 104% for green and red devices compared to the conventional Alq single-host devices, respectively.

2. Experiment

The device configuration studied is indium-tin-oxide (ITO)/NPB (60 nm)/ADN: Alq: dopant (20 nm)/4,7-diphenyl-1,10-phenanthroline (BPhen) (10 nm)/Alq (30 nm)/LiF (1 nm)/Al (200 nm). NPB, ADN: Alq: dopant, Bphen and Alq next to LiF are used as hole transporting, emitting, hole blocking and electron transporting layers, respectively. And DCJTB or N,N′-dimethyl-quinocridone (DMQA) is used as the dopant in emitting layer (EML). The device configuration and the chemical structures of materials in the EML are shown in Fig. 1. The ITO-coated substrates were routinely cleaned by ultrasonic treatment in solvents and then cleaned by exposure to a UV-ozone ambient. All organic layers were deposited in succession without breaking vacuum (3×10^{-4} Pa). Thermal deposition rates for organic materials, LiF, and Al were ~1, ~1 and ~10 Å/s, respectively. EL and photoluminescence (PL) spectra were measured with a Hitachi F-4500 fluorescence spectrophotometer. Absorption spectra were measured with a Shimadzu UV-3101 PC scanning spectrophotometer. The luminance–current–voltage characteristics were measured with a Keithley model 2400 power supply combined with an ST-900M spot photometer and were recorded simultaneously with measurements.

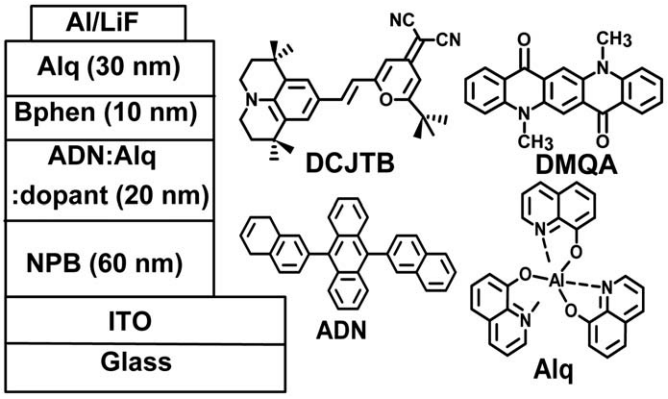

Fig. 1. The device configuration and the chemical structures of materials used in the emitting layer.

3. Results and discussion

Figs. 2(a) and (b) show the current efficiency as a function of the current density for a series of cohost-based OLEDs doped with 0.8 wt% DMQA and 2 wt% DCJTB, respectively. For the DMQA devices, a highest current efficiency of 15.14 cd/A is attained when the ADN/Alq ratio is 9:1, which increases by 54% and 48% compared to that of the Alq (9.86 cd/A) and ADN (10.21 cd/A) single-host devices, respectively. While the maximum current efficiency of the DCJTB devices is 6.05 cd/A when the ADN/Alq ratio is also 9:1, which increases respectively by 104% and 7% compared to 2.97 cd/A and 5.65 cd/A of Alq and ADN single-host devices. The details of the EL performances for the cohost devices doped with 0.8 wt% DMQA or 2 wt% DCJTB are listed in Table 1. Fig. 3 shows the EL spectra of the DMQA and DCJTB devices with different ADN/Alq ratios. It is noticed that the EL spectra shift to shorter wavelength with the increase in the ADN ratio in the cohost systems, so the Commission Internationale De L'Eclairage (CIE)-1931$_{x,y}$ coordinates vary too. The EL peaks of DMQA and DCJTB from each device in which the ADN/Alq ratios are 10:0 are almost the same with the PL peaks of DMQA and DCJTB shown in

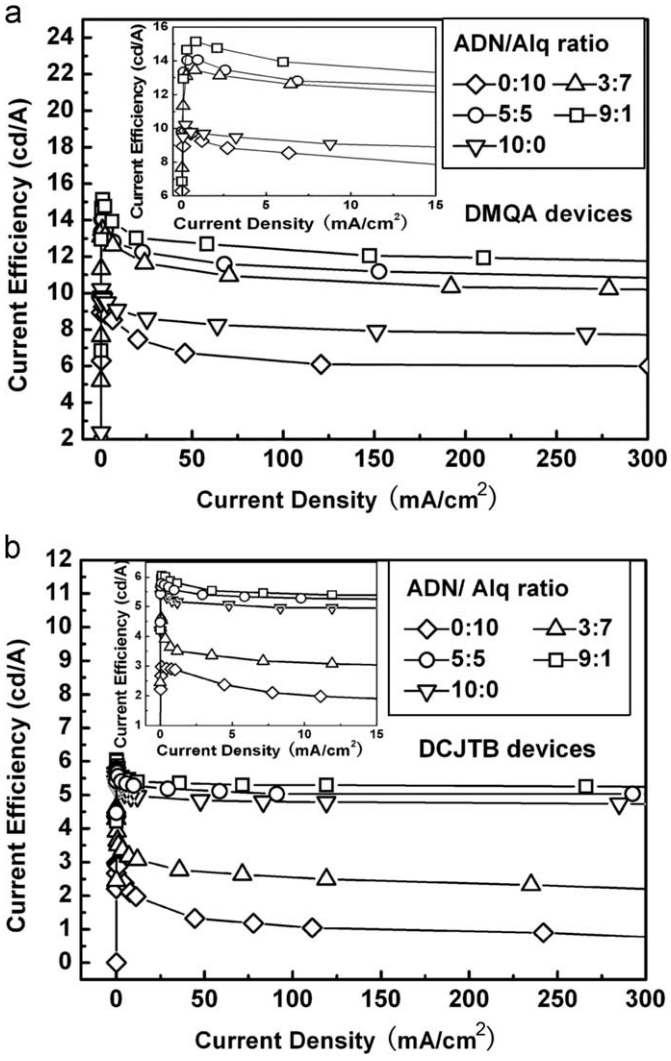

Fig. 2. The current efficiency–current density characteristics of the DMQA devices (a) and DCJTB devices (b) with different ADN/Alq ratios in the cohost. Insets: The enlargement of the current efficiency–current density characteristics of the initial ranges (0–15 mA/cm^2). The DMQA and DCJTB concentrations are the optimized concentrations by considering both the EL efficiencies and the CIE coordinates of the OLEDs.

Table 1
Details of EL performances of DMQA and DCJTB devices with different ADN/Alq ratios in the cohost at the current densities of 20 and 100 mA/cm^2.

Dopant	ADN/Alq ratio	J=20 mA/cm^2		J=100 mA/cm^2	
		η (cd/A)	CIE$_{x,y}$ coordinates	η (cd/A)	CIE$_{x,y}$ coordinates
DMQA (0.8 wt%)	0:10	9.0	(0.349, 0.635)	6.2	(0.335, 0.626)
	3:7	13.1	(0.338, 0.641)	9.6	(0.326, 0.631)
	5:5	13.6	(0.333, 0.646)	10.3	(0.321, 0.637)
	9:1	14.7	(0.334, 0.644)	12.2	(0.328, 0.635)
	10:0	9.6	(0.308, 0.605)	8.0	(0.301, 0.596)
DCJTB (2 wt%)	0:10	1.8	(0.645, 0.354)	1.2	(0.638, 0.362)
	3:7	3.4	(0.644, 0.356)	2.5	(0.636, 0.363)
	5:5	5.5	(0.644, 0.357)	5.0	(0.636, 0.365)
	9:1	5.7	(0.623, 0.375)	5.3	(0.620, 0.381)
	10:0	5.1	(0.594, 0.405)	4.8	(0.586, 0.410)

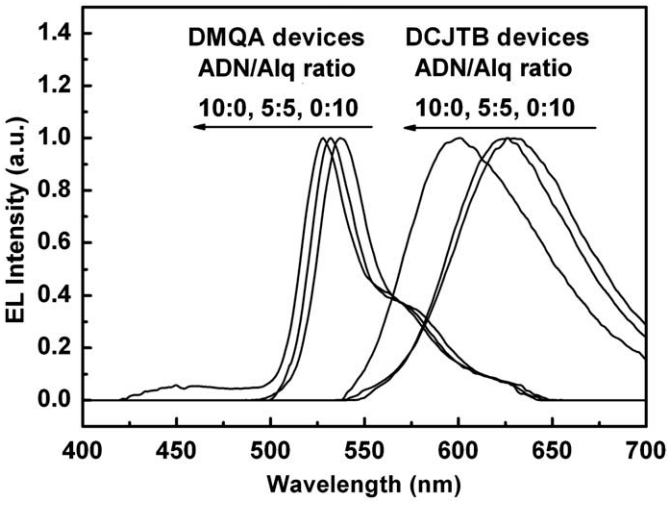

Fig. 3. EL spectra of DMQA and DCJTB devices with different ADN/Alq ratios in the cohost at the current density of 20 mA/cm^2.

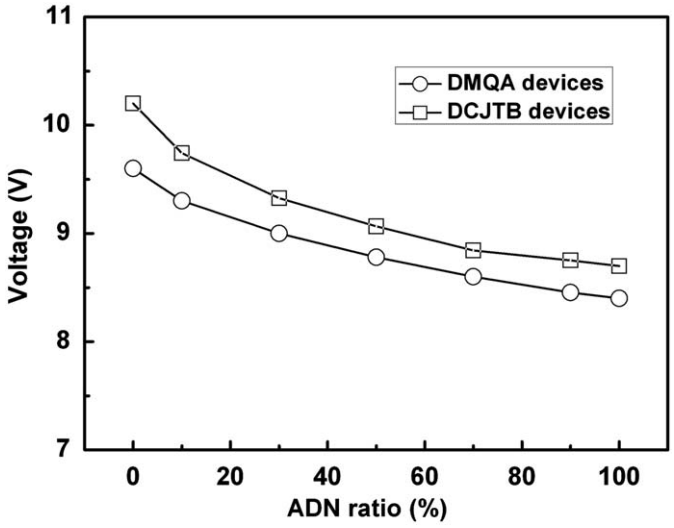

Fig. 4. The voltage–ADN/Alq ratio characteristics of the DMQA and DCJTB devices at the current density of 20 mA/cm^2.

Fig. 5(b). Therefore, the spectrum shift should be due to the solid-state solvation effect [8]. The DMQA devices provide a blue ADN emission when the ADN/Alq ratio is 10:0. This indicates that the energy cannot be completely used by the dopant when the concentration of the dopant is low.

Fig. 4 shows the dependence of the driving voltage on the ADN fraction at the current density of 20 mA/cm^2. We can see that the driving voltage decreases with the increase of the ADN fraction. It is proposed that this phenomenon is attributed to the good bipolar transporting character of ADN [9], which can lead to the balanced electrons and holes in the EML and the enlarged width of recombination region. Thus the cohost devices can achieve higher EL efficiencies.

Fig. 5(a) depicts the optical absorption and PL spectra of the materials used in this work. We note that the overlaps between the PL spectrum of ADN and the absorption spectra of DMQA and DCJTB are very small. This indicates that the energy transfer from ADN to DMQA or DCJTB may be inefficient. Fig. 5(b) reveals the PL spectra of the films of ADN doped with 0.8 wt% DMQA and ADN doped with 2 wt% DCJTB, respectively. It is obvious that the PL emissions of ADN in both the blend films are rather strong although the DMQA and DCJTB dopants also emit green and red emissions. However, the 0.8 wt% DMQA and 2 wt% DCJTB-doped ADN single-host devices exhibit much smaller or do not exhibit any blue EL emission, as plotted in Fig. 3. The differences in the ADN emission intensities between the EL and PL spectra ulteriorly imply that the EL emission mechanisms from DMQA and DCJTB are trapping in the DMQA- and DCJTB-based ADN single-host devices. However, large spectral overlaps among Alq, DMQA and DCJTB are observed in Fig. 5(a). Therefore, we can expect the Förster energy transfer from Alq to DMQA and DCJTB should be efficient. The PL spectra of ADN-doped Alq films are shown in Fig. 5(c). We can conclude that the energy transfer from ADN to Alq is complete under the 1:9–9:1 ADN/Alq ratios since there is no ADN emission in the PL spectra. According to the above experimental results, the emission processes in the DMQA- and DCJTB-doped ADN/Alq cohost devices can be interpreted as follows. As shown in Fig. 5(d), there are three possible emission processes in the ADN/Alq/dopant cohost systems:

(1) The injected carriers from the electrodes recombine on the ADN molecules and excitons are formed on ADN sites. The excitonic energy is then effectively transferred to Alq molecules. After that, the excitonic energy continues to transfer to dopants and finally produce the light from dopants.
(2) Injected electrons and holes recombine on the Alq molecules and the Alq molecules enter an excited state. The excitonic energy of Alq is then transferred to the dopants, finally the dopant molecules produce the light.
(3) Injected electrons and holes directly recombine on the dopant molecules, and the dopant molecules produce the light.

The three EL processes take place synchronously, and they compete with one another when the cohost devices are working. The competition of the three processes might make the carriers injected from the electrodes to be utilized more adequately

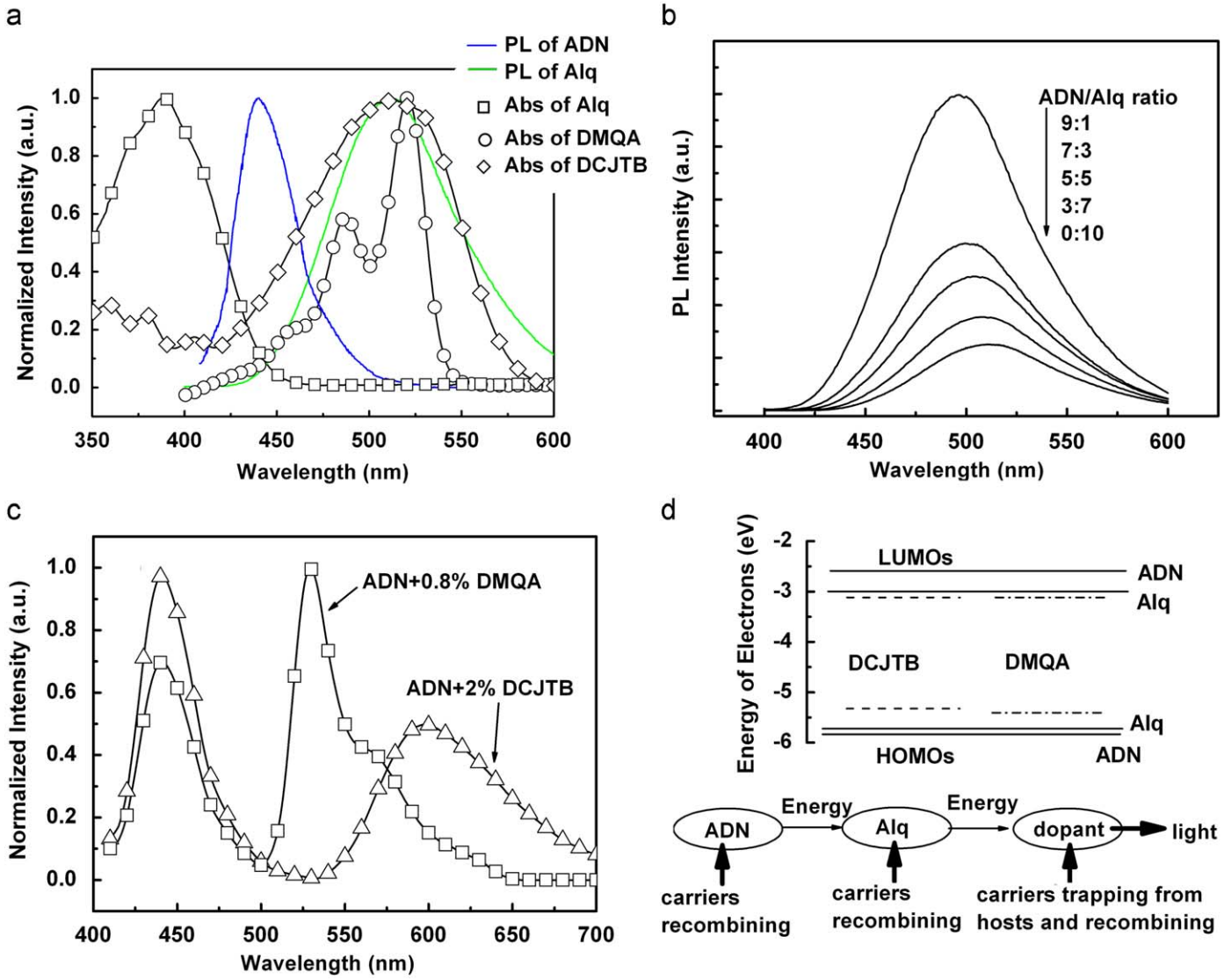

Fig. 5. (a) PL spectra of ADN and Alq in films and absorption spectra of Alq, DMQA and DCJTB in chloroform solution. (b) PL spectra of 0.8 wt % DMQA and 2 wt % DCJTB in ADN films. (c) PL spectra of different ADN/Alq ratio films. The blue shifts of the PL peak of Alq with decreased Alq ratio could be attributed to the 'polarization induced' solid-state solvation effects. The PL intensity of Alq increases is due to the non-radiative decay process of Alq retarded and the resulted PL efficiency of Alq increase [10]. (d) Energy diagram of EML and the sketch map of the emission processes of cohost devices.

[4,11,12]. As a result, the efficiencies of OLEDs are greatly improved.

It is worth noticing that the ADN and Alq cohost system can also be used as an emitting host for other green and red dopants to produce efficient green or red emission, such as C-545T, Coumarin 6 and 4-dicyanomethylene-2-methyl-6-(p-dimethylaminostyryl)-4H-pyran. This feature is extremely important and will be used to simplify the process of the solid-state lighting and full-color OLED fabrication.

4. Conclusion

In summary, we investigated an efficient ADN and Alq cohost system that can be used for green and red fluorescence dyes. The cohost devices show dramatically enhanced current efficiencies and decreased working voltage compared to the conventional Alq single-host devices. We attribute the increase of the efficiency to the balanced hole and electron injection into EML, the enlarged width of recombination region and the multiple emission processes. The cohost technique in this paper is expected to provide a valuable route to construct highly efficient fluorescence OLEDs.

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