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2002 Chinese Phys. Lett. 19 1697

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## Carrier Transport Behaviour of Molecular Doped Poly(N-Vinylcarbazole) in Polymer Light-Emitting Diodes \*

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(Received 22 May 2002)

Single-layer polymer light-emitting diodes are prepared from blends of poly(N-vinylcarbazole) (PVK) doped with tris(8-hydroxy-quinoline) aluminium (Alq<sub>3</sub>) of 2 wt% (sample a) and 0.2 wt% (sample b). The onset of PVK transient electroluminescence (EL) is delayed with respect to that of Alq<sub>3</sub> in sample a under pulsed excitation, while the EL onsets of Alq<sub>3</sub> and PVK in sample b are simultaneous. The total carrier mobility of the Alq<sub>3</sub>-rich regions in sample a is larger than that of the PVK-rich regions. However, the total carrier mobility is homogeneous in sample b. The phase image of atomic force microscopy and photoluminescence spectra of samples a and b indicate that the separated phase of samples a and b exists in the PVK-rich and Alq<sub>3</sub>-rich regions. The variance of the doping concentration and separated phase in blends results in the different carrier transport mobility of Alq<sub>3</sub>-rich and PVK-rich regions.

PACS: 78.60.Fi, 73.50.-h

Molecular dopants in polymer light-emitting diodes (PLEDs) are often used to enhance the brightness and efficiency of PLEDs and to realize multi-colour or white emission.<sup>[1-5]</sup> However, phase separation in nanometre to micrometre ranges is always formed spontaneously when doped polymer blends are spin-coated, due to the small entropy of mixing.<sup>[6]</sup> Phase separation is usually investigated by such techniques as electron scattering, atomic force microscopy (AFM) and near-field scanning microscopy.<sup>[6,7,11]</sup> The above techniques can only distinguish the topology and phase-separation scale of films, but cannot offer the electrical and luminescent information of blends. Here we make use of transient electroluminescence (EL) to study the carrier transport behaviour of blends at low doping concentration. Transient EL has been extensively applied in the research of carrier transport in single-layer and multi-layer organic LEDs.<sup>[8,9]</sup>

We prepared two blends of poly(N-vinylcarbazole) (PVK) doped with 2 wt% and 0.2 wt% Alq<sub>3</sub>, called samples a and b, respectively. The blend solutions were spin-coated on to indium-tin oxide (ITO) coated glass and then lithium fluoride (LiF) with 1 nm and aluminium, as a cathode, was evaporated in the vacuum pressure of  $2 \times 10^{-3}$  Pa. The thickness of the blends was about 80–90 nm. The active area of the diodes was 10 mm<sup>2</sup>. The luminescent spectra were measured by using a SPEX Fluorolog-3 spectrometer. The AFM phase images of the blends were investigated by using tapping-mode AFM (Quesant 250).

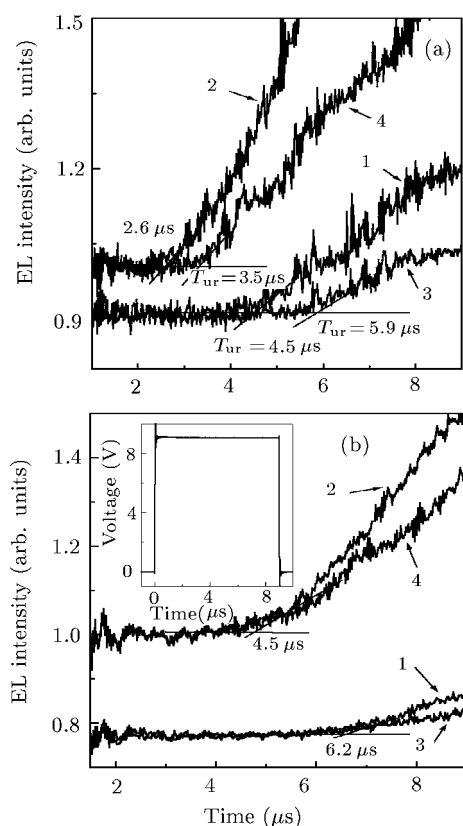
For the transient EL measurements, we applied voltage from an AV-1011-B (10 ns rise and fall time) pulse generator and collected emission using a photomultiplier (PMT) in the SPEX Fluorolog-3 spectrometer. The EL response and voltage were digitized by a Tecktronic TDS5400 oscilloscope. The resistivity-capacitance (RC) time constant of the system is estimated to be less than 0.1  $\mu$ s by  $T_r = 2.19$  (RC time constant),<sup>[10]</sup> where  $T_r$  is the transition of voltage leading and a trailing edge from 10% to 90%.

Figure 1 presents the transient EL behaviour of Alq<sub>3</sub> and PVK in PLEDs. The pulse length was 9  $\mu$ s, the repetition rate was 1 kHz and the EL response was averaged over 200 pulse periods. To compare different EL signals, the stable baselines are normalized. The EL onset times of Alq<sub>3</sub> ( $T_{tra}$ ) and PVK ( $T_{trp}$ ) in sample a are about 4.5  $\mu$ s and 5.9  $\mu$ s at 10 V, and the delay times are 2.6  $\mu$ s and 3.5  $\mu$ s at 12 V, respectively (Fig. 1a). With the increasing voltage pulse, the EL onset delay times decrease and the EL onset delay time difference ( $\Delta T = T_{trp} - T_{tra}$ ) detracts. However,  $\Delta T$  exists even in a high voltage pulse (18 V) (not shown). The EL responses of Alq<sub>3</sub> and PVK are simultaneous, even at the threshold voltage (9 V) and their delay time is about 6.2  $\mu$ s. The quick pulse voltage in the inset of Fig. 1(b) shows that the EL response of the PLEDs is determined by the product of carrier mobility and electric field, not by the RC of the system. The results suggest that the carrier transport behaviour of the blends varies with the doping concentration.

To research further reasons for different carrier

\* Supported by the National Natural Science Foundation of China under Grant Nos 29992530, 19974002 and 29771029, and the Foundation of Northern Jiaotong University.

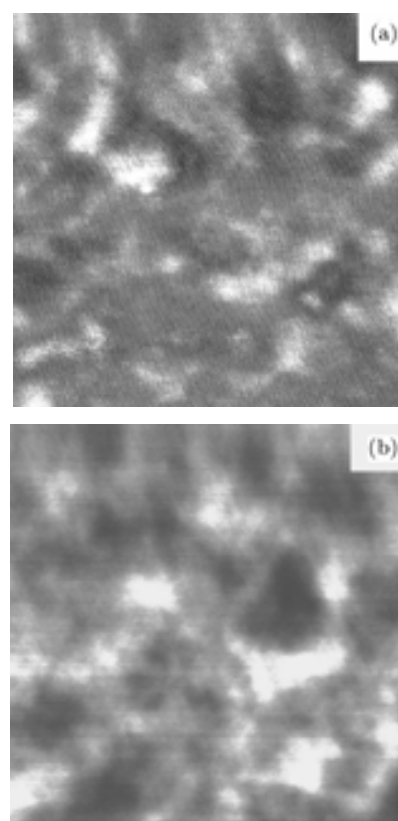
transport behaviour, we study the AFM phase images of Alq<sub>3</sub> doped PVK blends. The tapping-mode AFM images of the blends are shown in Fig. 2. The phase separation can be detected in both sample *a* (Fig. 2(a)) and sample *b* (Fig. 2(b)). The isolated phase domains shows a cloud-like structure of 60–140 nm diameters in the blends, while the phase domains of the phase image in sample *b* are more outstretched and faint than those of sample *a*. Why does not it attain  $\Delta T$  in sample *b*? The reason is that the AFM technique cannot discern more precise difference due to its resolution limit.<sup>[11]</sup> The spectral analysis of electroluminescence (EL) and photoluminescence (PL) probes<sup>[6]</sup> can provide a further signature of phenomenon, which is not detected by AFM.



**Fig. 1.** EL response of Alq<sub>3</sub> and PVK in PLEDs blended with 2 wt% (sample *a*) and 0.2 wt% (sample *b*) Alq<sub>3</sub> in PVK blends at rectangular voltage pulses. (a) Delay time of Alq<sub>3</sub> [(1) at 10 V and (2) at 12 V] and PVK ((3) at 10 V and (4) at 12 V) in sample *a*. (b) Delay time of Alq<sub>3</sub> [(1) at 9 V and (2) at 12 V] and PVK ((3) at 9 V and (4) at 12 V) in sample *b*. The EL signal monitoring peaks of Alq<sub>3</sub> and PVK are located at 500 nm and 410 nm. The inset is the pulse voltage of 9 V.

Figure 3 presents the EL and PL spectra of blends and pure Alq<sub>3</sub> films. The peaks of spectra are blueshifted from 513 nm to 495 nm when the Alq<sub>3</sub> concentration in the blends decreases gradually. The peaks of PL and EL spectra from sample *a* are redshifted with respect to those of sample *b*. The lu-

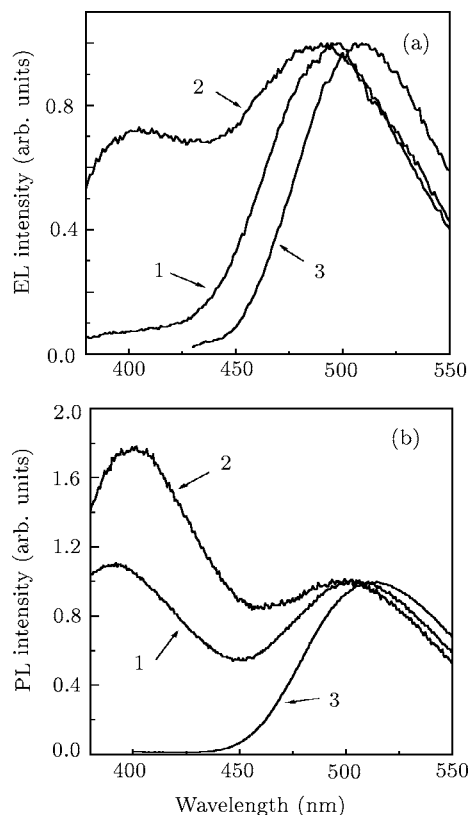
minescent intensity ratio of PVK and Alq<sub>3</sub> increases with the decrease of Alq<sub>3</sub> doping concentration due to the effect of the Förster-type energy transfer.<sup>[6,12]</sup> Depending on the AFM phase images and PL and EL probes, the phase separation of the blends exists in Alq<sub>3</sub>-rich and PVK-rich regions. The interaction of Alq<sub>3</sub> molecules in the PVK matrix in sample *b* is much weaker and the Alq<sub>3</sub> inter-molecular distance in sample *b* is larger than that of sample *a*.



**Fig. 2.** Tapping-mode AFM images of PVK blends doped with 2 wt% (a) and 0.2 wt% (b) Alq<sub>3</sub>. The images are shown in phase contrast with an area of  $1 \times 1 \mu\text{m}^2$ .

In principle, the total mobility ( $u_t$ ), which is the summation of hole mobility ( $u_h$ ) and electron mobility ( $u_e$ ), is estimated by  $u_t = D/(T \times E)$ , where  $D$  is the effect thickness of blend layer,  $T$  is the delay time of transient EL and  $E$  is the electric field. The  $u_e$  value of pure Alq<sub>3</sub> films is in the range of  $10^{-6}$ – $10^{-5} \text{ cm}^2/\text{Vs}$ <sup>[9]</sup> and that of pure PVK films is  $10^{-7}$ – $10^{-6} \text{ cm}^2/\text{Vs}$  at the electric field ranging from  $1.5 \times 10^5$  to  $3 \times 10^6 \text{ V/cm}$ .<sup>[13]</sup> However, the values of  $u_e$  of Alq<sub>3</sub> and  $u_h$  of PVK in the diluted films decrease according to the relation of  $u \sim \exp(-d)$ .<sup>[14]</sup> According to Figs. 2 and 3, the hopping between Alq<sub>3</sub> molecules in Alq<sub>3</sub>-rich regions occurs in sample *a*. Therefore, in Fig. 1(a)  $\Delta T$  of sample *a* is attributed to different  $u_t$  of Alq<sub>3</sub>-rich and PVK-rich regions. While the Alq<sub>3</sub> molecule interactions in Alq<sub>3</sub>-rich regions in sample *b* are very weak, the hopping rate between Alq<sub>3</sub> molecules is relatively low in sample *b*. The Alq<sub>3</sub> molecules are isolated

in the PVK matrix and Alq<sub>3</sub> act as carrier traps in sample *b*.



**Fig. 3.** Luminescent spectra of Alq<sub>3</sub> doped PVK blends and Alq<sub>3</sub> pure film. The EL (a) and PL (b) spectra of PLEDs with (1) 2 wt%, (2) 0.2 wt% Alq<sub>3</sub> doped PVK blends and (3) Alq<sub>3</sub> pure film.

Hence, the EL of Alq<sub>3</sub> results from the energy transfer of PVK or the recombination of the trapped electron of Alq<sub>3</sub> and the transported hole of PVK. As a result, the onset time of Alq<sub>3</sub> EL is decided by the mobility of PVK blends.

In summary, we have studied the onset of Alq<sub>3</sub> EL and that of PVK in PLEDs with different doped concentrations using transient EL. Experiments show that the separated phase in sample *a* results in faster mobility in Alq<sub>3</sub>-rich regions while the effect of Alq<sub>3</sub> traps in sample *b* causes similar mobility between Alq<sub>3</sub>-rich and PVK-rich regions. The doping concentration and separated phase result in the carrier mobility variance in the space distribution.

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