

## Enhanced energy transfer between poly-(vinylcarbazole) and tris(2-phenylpyridine) iridium(III) via the switch of molecular conformation induced by different solvents and UV light irradiation

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### ABSTRACT

In this letter, we reported the influence of solvents or UV light irradiation on EL efficiency of phosphorescence-doped PLEDs. The molecular conformation of host material PVK depended on different treatment process, which can be switched from side-by-side structure to face-to-face structure. More triplets are formed inside PVK molecular with face-to-face conformation structure that induced lower carrier transport ability. Therefore, it is significant to optimize the treatment process of phosphorescence-doped PLED to achieve higher efficiency by molecular conformation controlling.

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The conformation of polymer molecules has significant effect on its optoelectronic properties. It has attracted an extensive research interest to investigate its influence on device performance of polymeric optoelectronic devices such as LED or solar cell [1–5]. Among them, one of very interesting phenomena is that molecules conformation has effect on triplet formation probability [6,7]. Generally, people can control the molecules conformation of polymer via some technique methods such as thermal or vapor, treatments of as-cast films or by spinning films from solvents with different parameters as boiling points and solubility [8–10,20,21].

PVK, as a classic non-conjugated polymer, attracted great research attentions due to its good holes transport ability and excellent candidate for phosphorescence-doped PLEDs. The main luminescent emission of PVK comes from an excimer caused by two adjacent flat carbazole units stacking relative to one another [13,14]. A lower energy excimer (420 nm) is formed when two carbazole groups form a fully eclipsed “sandwich-like” conformation. This is so called face-to-face structure. A second higher energy excimer state (380 nm) results from a partially eclipsed conformation that involves only one eclipsed aromatic benzene ring from each carbazole group (side-by-side structure).

In our previous works, it is found that molecules conformation of PVK molecule mostly exist in side-by-side structure in film spin-coated from chlorobenzene, while that from chloroform mostly exist in face-to-face structure [15]. Furthermore, we confirmed that the two molecular conformation ratio of triplet to singlet excitons is different. Firstly, the face-to-face conformation has a larger triplet/singlet ratio compared to the side-by-side conformation and secondly, most free polarons from the dissociation of singlet exciton re-associate to form triplet excitons due to spin statistics. So the formation probability of triplet excitons in the face-to-face conformation is higher than that in the side-by-side conformation.

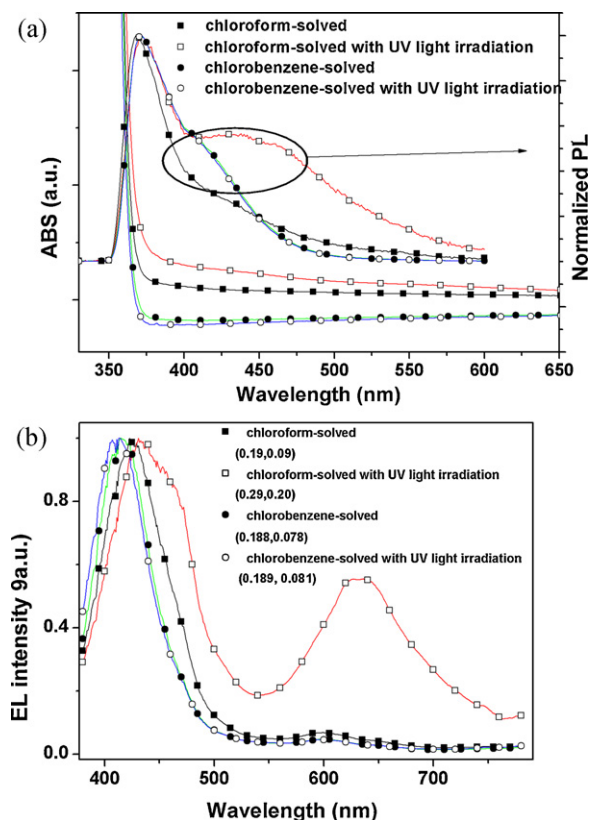
On the other hand, photo-physical and photo-chemical have significantly effect on molecular structures of polymer as well as optoelectronic properties. Moreover, polymer molecules in different conformation have different photo-physical and photo-chemical activity. Feringa et al. obtained reversible three-state switching of luminescence (blue–nonfluorescent–red) through the subtle interplay of photochemical and electrochemical stimuli [11]. Ji and Jiang increased the electrical conductivity of poly(vinylidene fluoride) by excimer laser irradiation with  $\lambda = 248$  nm [12].

In this letter, we fabricated phosphorescence-doped PLEDs based on PVK film spin-coated from chloroform or chlorobenzene with and without irradiated by UV light ( $\lambda = 365$  nm) based on the above results. The different effect of UV light irradiation on PVK in chloroform and chlorobenzene was compared, which caused by different molecules conformation. In addition, the device performance

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**Fig. 1.** (a) Absorption and PL spectra of PVK solution in different solvents and with or without UV light irradiation. Their EL spectra are shown in (b) at 10V. The inset is the corresponding CIE data of these EL spectra.

of phosphorescence-doped PLEDs based on PVK from chlorobenzene enhanced much significantly than that from chloroform.

The EL device consisted of ITO/PEDOT-PSS/PVK/Ca:Al multilayered structure was investigated. The 30 nm thick poly(3,4-ethylenedioxythiophene) poly(styrenesulfonate) (PEDOT-PSS) layer was spin-coated onto a cleaned glass substrate coated with indium tin oxide (ITO) and then annealed at 150 °C in vacuum for 4 h. The 100 nm thick PVK layer was spin-deposited on the annealed PEDOT-PSS layer. PVK (purchased from Aldrich, molar weight of 90,000 was used without further purification) was dissolved in chloroform or chlorobenzene at 10 mg/ml except where reported otherwise. Then the films were annealed in vacuum at 180 °C for 1 h. 5 nm thick Ca and 200 nm thick Al contact, 3 mm in diameter, were vapor deposited onto the PVK layer to complete the device. For dye-doped EL devices, the weight ratio of phosphorescent dyes (tris(2-phenylpyridine) iridium(III)) in PVK host was 0.8 wt%. Photoluminescence (PL) spectra were collected using a monochromatized Xe light source (300 W). EL emission was collected by a fiber optic and dispersed onto a CCD Si detector (Ocean Optics Spectrometer). The current–voltages data were measured using a Keithley 238 power source.

Absorption and PL spectrum of PVK in different solvent with and without UV light irradiation was shown in Fig. 1a. The absorption spectra of chloroform-solved PVK red-shift compared with that of chlorobenzene-solved PVK, which implied chlorobenzene have better solubility to PVK than chloroform. More coiled and tightly PVK molecular in chloroform induced PVK chain stacking together, which actually increased aggregation states and making absorption red-shift. After UV light irradiated, absorption spectra of chloroform-solved PVK extended to lower energy region whereas that of chlorobenzene blue-shift a little bit. Generally, there are four different photo-induced physical and chemical procedures

happened in PVK solution during UV light irradiation. Procedure 1: photo-induced polarization of carbazole groups, which formed more face-to-face structure; procedure 2: photochemical reactive, which cause crosslink between neighbor PVK molecular via bridge oxygen atom; procedure 3: photo-scission process and procedure 4: individual carbazole group broken down from PVK main chain by UV light irradiation. All of them could cause the polymeric length of PVK chain decreased. So we suggested that procedure 1 and 2 happened fast in chloroform-solved PVK, which made polymeric length of PVK chain and stacking degree of carbazole groups increased and then absorption spectra red-shift. As we know, more carbazole groups in face-to-face structure in chloroform-solved PVK while more carbazole groups in side-by-side structure in chlorobenzene-solved PVK [15], it means that different conformation have different influence on these four different photophysical and photochemical procedure. Face-to-face structure has larger overlap between neighbor carbazole compared with side-by-side structure, so it has relative lower intramolecular reorganization energy that arises from the change in equilibrium geometry of the donor and acceptor sites consecutive to the gain or loss of electronic charge upon electron transfer. Therefore, PVK with more face-to-face structure is easier to polarization under photoexcitation than that with more side-by-side structure. On the other hand, due to the different chemical properties of solvents as well as its influence on the conformation of PVK chain, the rate of photochemical procedures of PVK is quite different in different solvents. For PVK with more side-by-side structures, it should be very easy to be happened for photo-scission due to the higher reorganization energy, which made geometric change difficult and caused chemical bond broken. In PL spectra, chloroform-solved PVK no matter which is irradiated by UV light or not show stronger singlet excimer emission from face-to-face structure (~420 nm) than that of chlorobenzene-solved samples. And after UV light irradiation, this lower energy singlet excimer emission became stronger. It is possible because the carbazole groups in side-by-side structure changed to face-to-face structure under UV light irradiation.

Since PL properties of PVK depended on their molecules conformation, as the face-to-face structure for the lower energy excimer emission and the side-by-side structure for the high energy excimer states. The conformation of PVK molecular is changed in different solvents as well as luminescent properties. Moreover, the different conformations have different effect on photophysical and photochemical properties. The lower energy emission increased more obviously in chloroform-solved PVK than in chlorobenzene-solved PVK. It means that the photophysics phenomena such as polarization to face-to-face structure is easier happen in former one than in latter one.

The structure of single EL devices is as following: ITO/PEDOT:PSS/PVK/Ca:Al. All these EL devices showed two different features in EL spectra (Fig. 1b): 410–420 nm from singlet excimer emission and 590 nm from triplet excimer emission. In addition, it can be seen that the singlet excimer emission from chloroform-solved PVK red-shifted compared with that from chlorobenzene-solved PVK, which implied more face-to-face structure in former one. It is confirmed that the formation probability of triplet excitons increased in PVK molecules with face-to-face conformation compared with that with side-by-side conformation [15]. So the relative intensity of triplet emission is higher in chloroform-solved PVK than that in chlorobenzene-solved PVK. With UV light irradiation, the triplet excimer emission increased more in chloroform-solved PVK than in chlorobenzene-solved one. As we mentioned above, PVK film with more face-to-face structure is easier polarization, which caused much more face-to-face structure under high electric field and then increased triplet excimer emission. On the other hand, the singlet excimer emission from face-to-face structure ~460 nm enhanced after UV light irradiation,

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