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Post-functionalized poly(*N*-vinylcarbazole)s as effective low turn-on-voltage phosphorescent-hosts in polymeric light emitting devices

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ABSTRACT

DTAB-PVKs are prepared by post-functionalization of poly(*N*-vinylcarbazole) (PVK) with di(4-tolyl)aminobenzyl acetate (**DTABOAc**) as the pendant groups via the Friedel–Crafts reaction. Introduction of the methyl substituents at the *para*-position of **DTABOAc** is essential for suppression of its self-condensation. The details of the reactions mechanisms are elucidated by ¹H NMR study. The Fermi level of **DTAB-PVKs** could be tuned by controlling the amounts of **DTABOAc** content in the polymer and **DTAB-PVKs** are found to be good hosts for phosphorescent polymer light-emitting diodes (PLEDs). In a device of PLED comprising PEDOT: PSS/**DTAB-PVK**-Ir(ppy)₃-PBD/Mg–Ag, low turn on voltage at 6.5 V is found with a reasonably high current efficiency of 11.5 cd/A at 13.0 V. The power efficiency reaches 2.7 lm/W at 12.0 V. More importantly, the device performance related to the Fermi level of **DTAB-PVKs**.

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1. Introduction

Polymer light-emitting diodes (PLEDs) have been a subject of intense research over the years because of their potential uses in the display industry [1]. The Fermi level and the charge transport properties of a polymer are important factors that govern the performance of the PLEDs. To construct an efficient PLED, charge injection and transport properties have to be tuned so that the hole–electron recombination could be confined at the emissive zone that is known as the recombination zone. The location of the radiative charge recombination strongly affects the

performance of the PLED. For example, recombination zones that are located within a short distance from the electrode surface should be avoided. It is because the metallic structure provides a non-radiation decay channel, which may be responsible for the quenching of the emitting layer [2–6], leading to low device efficiency [7–10]. Therefore, multilayer PLED devices are designed to deal with this problem; the PLED devices containing: hole-transporting layer, light emitting layer and electron-transporting layer become major target to study. In particular, the design of the light emitting layer is crucial for optimization of the device efficiency.

In the past few years, many research teams have put efforts in exploration of the molecular design to control their electronic properties, for examples, by changing the conjugated length of the molecule [11] or by physical blending [12]. Although the method of using physical blends of two or more materials to form a single layer provides a convenient way for preparing optoelectronic films, phase

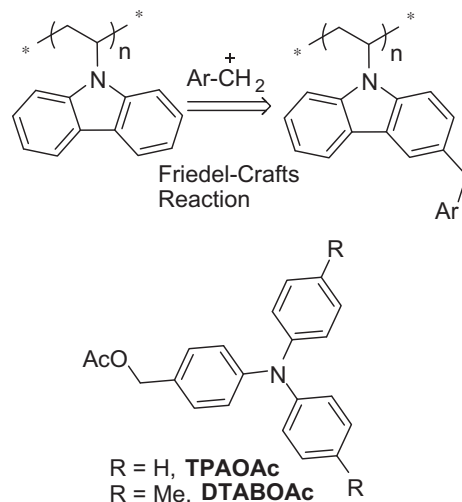
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separation of the components may occur and cause defect in the device and current leakage. Therefore, the preparation of single polymers with components equipped on the polymer chain is desirable.

To be a good phosphorescent host-matrix, the material having high-energy triplet is an essential criterion to fulfill. The triplet energy of the host should be higher than that of the triplet emitter so that the triplet exciton-energy back transferring from the triplet emitter to the host matrix could therefore be prevented. For example, poly(*para*-phenylene-(*E*)-vinylene) [13] (PPV) and poly(3,6-fluorene) (PF) [14] have a maximum triplet energy of 1.6 and 2.4 eV, which make their application suitable as host polymers for red and yellow triplet emitters respectively.

To design host polymers with high triplet energy for phosphorescence PLED, the first factor was to control the HOMO/LUMO gap that has to be large enough to fit for triplet emission. In addition, the HOMO/LUMO energy levels should match well with the adjacent layers in order to maintain efficient charge injection and transport properties. Moreover, polymer synthesis has to be simple and efficient to fulfill the mass production criteria. Furthermore, it would be better to use transition-metal free synthetic process in order to avoid luminescence quenching by any residual contaminants. All these are challenging problems to deal with. Besides, to enhance the power efficiency of the PLED, device having low turn-on voltage is preferred. Poly (9-vinylcarbazole) (PVK) is a well known photo-conductor with superb electronic properties and high triplet energy [15–17,7,18,19]. However, due to the mismatched HOMO level with that of ITO, the hole-injection barrier for the PLED is usually high. Therefore, a new generation of PVK based materials is highly desired. Since PVK has reasonable chemical reactivity towards electrophilic aromatic substitution, PVK could be easily post-functionalized in few steps [20,21]. The most of important is the fact that PVK is commercially available and could be obtained in large quantity. All these make PVK a good candidate to study. During the past decades, many efforts have been investigated in post-functionalization polymer field which were applied in optical [22–24], electronic [25], drug delivery [26], and surface modification [27]. The approach of post-functionalization provides an opportunity for the systematic investigation on the photophysical, electronic and thermal properties of a polymer system, without being bothered by the tedium of monomer and polymer syntheses, and any uncertainty arising from variation of the degree of polymerization in the polymer synthesis could be avoided. In our previous paper [28], we demonstrated the use of triphenylamine (TPA) derivatives as a cross-linker for the PVK matrix to form a hole-transport layer. In addition, introduction of the TPA component could significant modify the properties of the PVK matrix; correlation between the amounts of triphenylamine components and the improvement of the PLED efficiency have been discovered. Therefore, we are interested in demonstrating the possibility of using PVK as the precursor for post-functionalization. The idea is illustrated in Scheme 1. Generation of the benzylic carbocations in the presence of PVK would lead to electrophilic substitution at the 3-, 6-positions.



Scheme 1. The idea of post-functionalization of PVK through the Friedel-Crafts reaction.

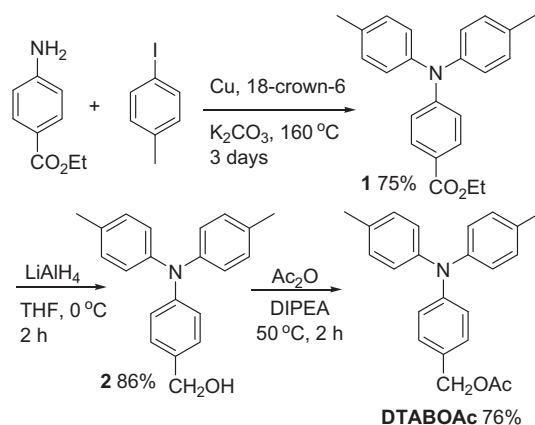
In this study, we report a novel and simple approach of modifying PVK with TPA pendant groups such as di(4-tolyl)aminobenzyl acetate (**DTABOAc**) through post-functionalization with the Friedel–Crafts electrophilic aromatic substitution reaction. This modification leads to reduction of the device turn-on voltage as well as the enhancement of the device power efficiency.

2. Results and discussion

2.1. Preparation of DTABOAc and TPAOAc

DTABOAc was prepared, as shown in Scheme 2, from the commercially available ethyl 4-aminobenzoate and 4-iodotoluene through the Ullmann coupling, followed by the LiAlH_4 reduction of **1** in THF to afford **2** that was further acetylated with acetic anhydride to give **DTABOAc** in good yields (76%).

TPAOAc was prepared according to literature procedures [28], by monoforylation of TPA, followed by NaBH_4



Scheme 2. Synthesis of **DTABOAc**.

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