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Review

Carbazole-based polymers as hosts for solution-processed organic lightemitting diodes: Simplicity, efficacy

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ABSTRACT

Polymer light emitting diodes (PLEDs) have attracted a great deal of interest within academia and industry because of their potential applications in flat panel displays and solid-state lighting technologies. The solution processability of polymers offers the advantages of simple and mild fabrication conditions enabling to cut cost and produce large area displays. Among all polymeric hosts under investigation, carbazole-based materials benefit from the wide bandgap of carbazole as well as its remarkable thermal, photochemical and chemical stability. Especially, the relatively high triplet energy level of carbazole makes it an appealing candidate to design hosts for wide bandgap triplet emitters such as blue dopants. In this review, an overview of all carbazole-based polymeric hosts reported to date is presented. Noticeably, easiness of synthesis has been an aspect largely developed to access to these polymeric structures.

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1. Introduction

During the past decade, great efforts have been made to develop highly emissive devices. Among the many different kinds of light-emitting organic materials available, phosphors (also called triplet emitters) are significantly superior to fluorophors. Indeed, device performances are intimately linked to the internal quantum efficiency (IQE) that can reach the light-emitting material used to fabricate organic light-emitting diodes (OLEDs) and phosphors clearly outperform fluorophors. Notably, a device emitting from a fluorescent material is limited by the theoretical ratio of singlet excitons (25%), thus constituting a severe internal drawback. However, the efficiency of fluorescent molecules has be recently increased to near the maximum of 100% by use of Thermally Activated Delayed Fluorescence (TADF) emitters [1]. As specificity, these materials offer a simple and robust method for boosting the efficiency obtained from traditional fluorescent emitters past the theoretical barrier of 25% to near 100% while exhibiting the high operational stability of fluorescent materials. But, the availability of these third generation emitters is still limited [2]. On the opposite, phosphorescent materials are more traditional emitters and these latter can harvest both electrogenerated singlet (25%) and

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triplet (75%) excitons for light emission, enabling to theoretically achieve 100% IQE [3-6]. Among all phosphors that have been investigated to date, iridium complexes have undoubtedly been the most widely studied. These complexes are highly versatile since neutral [7-12], cationic [13-17], anionic [18,19] but also soft salts [20,21], in which both the ion-paired anionic and cationic iridium complexes can participate to light emission, were successfully used as triplet emitters for OLEDs. These complexes also benefit from several appealing features including high quantum yield of luminescence [22,23], phosphorescence at room temperature [24] and relatively short excited state lifetimes so that key adverse factors such as triplet-triplet annihilation and back energy transfer from the guest to the host are strongly limited [25–28]. Emission color can also be easily tune by mean of the ligands introduced in the coordination sphere of the metal cation, enabling the emission to range from blue to red [29–35]. Beyond the simple control of the emission color, the emissive layer of an OLED comprising a triplet emitter cannot be only composed of a neat film of this latter (excepted for the emitters of specific design such as the dendrimers-like complexes) [36,37], but requires the metal complex to be diluted in a material named host to avoid self-quenching of the emitter [38]. To date, the most efficient devices are vacuum-processed and for the reproducibility of the electroluminescence (EL) performances, a precise control of the doping concentration of the injection/transport layers and/or the emissive layer during device fabrication are required.





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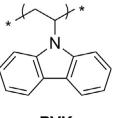
Performances thus strongly dependent of the control level achieved during the deposition process. Alternatives to vacuum-processed devices are thus actively searched. In this field, solution-processed OLEDs offer numerous advantages including a great ease of fabrication, reduced manufacturing costs and the potential of designing flexible and large area displays [39-43]. However, solution-processed devices require materials that are different from those commonly used for vacuum-processed devices as these materials are often small molecules. As drawback, small molecules exhibit a strong propensity to crystallize, constituting the main impediment to design solution-processed OLEDs with these molecules. This issue was partly addressed by the introduction of alkyl chains in the scaffold of molecular hosts even if the presence of non-conductive groups at high level can adversely affect charge transport [44]. On the opposite, polymers exhibit several advantages over small molecules such as a high solubility, a remarkable film-forming ability and a good thermal stability [45,46]. To make devices, one constraint on host choice relies in the fact that this material must exhibit a bandgap wider than that of the emitting material for an effective energy transfer from the host to the guest [47-49,4,50-52]. Especially, when triplet emitters are used, the energy level of the lowest excited triplet state (T_1) of the host must be higher than that the T_1 of the guest. Tremendous efforts are thus devoted to prepare wide bandgap polymeric hosts. However, availability of suitable polymeric hosts for blue phosphorescent dopants is still lagging far behind that of the other colors [53–55]. Recently, several examples of wide bandgap polymeric hosts based on a π -conjugated interrupted polymer backbone were developed [56–59]. Indeed, one major drawback of the conjugated main-chain polymers is their low bandgaps resulting from the π -electron delocalization along the polymer backbone. Without sacrificing the film-forming ability, non-conjugated main-chain polymers efficiently confine the π -electron delocalization within the repeating units, enabling to keep the bandgap of their individual units. In this field, one representative example is the well-known poly(*N*-vinylcarbazole) ($E_g = 3.5 \text{ eV}$) for which the polymer bandgap is similar to that of the carbazole moiety [60– 65]. Carbazole is among the most stable wide bandgap molecules that was extensively studied for the design of low molecular weight or oligomeric hosts [66], fully justifying the development of polymeric host with this molecule. As appealing features of PVK, this polymer exhibits a high glass transition temperature (200 °C), a high triplet energy level (E_T = 2.5 eV), a good solubility in common organic solvents and excellent film-forming properties, rendering this polymer of great interest for OLEDs [67–70]. However, PVK is characterized by a hole-dominated transportation, limiting its scope of usability [71,72]. As other drawback, PVK is also prone to exciplex formation, lowering device performances [73]. Development of carbazole-based polymers for solution-processed OLEDs with a more balanced charge transportation, high triplet energy levels and exhibiting HOMO and LUMO levels correctly matching those of the adjacent layers was not an easy task and this challenge was successfully overcome by chemists.

In this review, all carbazole-based polymeric hosts that have been reported to date are presented. Noticeably, materials design and evaluation in devices have been regarded as inseparable processes for a systematic approach to materials development. A close attention has been paid to simplify as much as possible the molecular design of these materials as well as the number of steps required to synthesize them. Benefiting from the wide bandgap of carbazole, efficient solution-processed blue polymer LEDs (PLEDs) could be prepared. By designing easily accessible materials, promising carbazole-based polymeric hosts were obtained and the polymers can be divided into two main families, namely those comprising carbazoles in the main chain and those bearing carbazoles as pendant groups.

2. PVK, a best seller as polymeric host

As already mentioned in the introduction, PVK has been extensively regarded as a polymeric host for PLEDs (see Fig. 1) [74–76], and it still constitutes a reference for evaluating device performances of the newly synthesized carbazole-based polymeric hosts. PVK is a unipolar hole transporter so that it must be blended with a large amount of electron transport materials to achieve a balanced charge carrier transportation [77]. Precisely, the hole mobility of PVK is about 3 orders higher than that of its electron mobility [63,64]. As electron-transport materials classically blended with PVK, 1,3,4-oxadiazole-2,2-(1,3-phenylene) *bis*[5-[4-(1,1-dimethylethyl)phenyl]] (OXD-7) [78,79] or 1,3,5-tris(N-phenylbenzimiazole-2-yl)benzene (TPBI) [80] are ones of those. Due to the imbalanced charge transportation, PVK-based devices require high driving voltages to operate but also underperform with blue dopants [71,72]. As origin of this counter-performance, PVK forms lower energy triplet dimers that quenches dopant emission and drastically reduces [81-88]. device efficiencies While using iridium(III) [*bis*(4,6-difluorophenyl)pyridinato- N,C^2]picolinate (FIrpic) as the dopant, the maximum power efficiency (PE) only reaches 0.8 lm/W [62]. By blending PVK with electron-transport materials, major improvements could however be achieved and blue PLEDs with peak current efficiencies ranging between 15 and 24 cd/A were obtained [82,89–92]. Extending these results to other emission colors proved to be a less challenging task. Indeed, current efficiencies reaching up to 40 cd/A could be measured with red and green dopants without blending PVK with electron-transport materials [93,94]. In the same spirit, white OLEDs could be fabricated by combining the blue phosphorescence of FIrpic with the red fluorescence of a silole and a carbazole-based copolymer PCz-MPTST [95]. A white electroluminescence with Commission Internationale de l'Eclairage (CIE) coordinates of (0.37, 0.40) and a current efficiency (CE) of 9.2 cd/A were obtained. Beyond the unipolar character of PVK, the low lying highest occupied molecular orbital (HOMO) of PVK (5.9 eV) that clearly mismatches the workfunction of common anodes is another major problem resulting in difficult hole injection from the anode (see Fig. 2).

The insufficient energy level of the triplet state of PVK ($E_T = 2.5 \text{ eV}$) is often invoked to support the low light-emitting efficiency of PVK-based devices, especially when deep blue dopants are used [96,97]. Nevertheless, highly emissive OLEDs can be obtained with the 4,4'-bis(N-carbazolyl)-2,2'-biphenyl (CBP):FIrpic system even if the E_T of this host is a little lower than that of the phosphor [98]. External quantum efficiencies (EQE) ranging from 6% to 14% were reported with this material. Therefore, to pay only attention to the triplet energy level of a host to anticipate performances is reductive and the control of the interfacial nature at the emissive layer (EML)/electron transport



PVK Fig. 1. Structure of PVK.

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