

Single white polymers based on simple diarylmaleimides: Polymeric structure and electroluminescent properties



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ABSTRACT

Two kinds of novel single white polymers, lateral-chain type and star-like type, were designed and synthesized by introducing di(*p*-methoxyphenyl)maleimide ($\lambda_{em} = 540$ nm) or dithienylmaleimide ($\lambda_{em} = 558$ nm) guest into the side chain of linear polyfluorene (LPF-TMs and LPF-PMs), and diphenylmaleimide ($\lambda_{em} = 500$ nm) guest into the core of star-like polyfluorene (SPF-PMs). By adjusting the feed ratio of maleimide and fluorene units, the resulting single white polymers exhibited a dual emission including blue emission (ca 430 nm) from polyfluorene host and yellow (ca 536 nm) emission from diarylmaleimide guest. Their electroluminescent properties were investigated in the single-emitting-layer devices fabricated by solution process with the configuration of ITO/PEDOT:PSS/polymer/TPBI/LiF/Al. Compared with main-chain type polymers, LPF-PMs and SPF-PMs displayed better electroluminescent performance with the maximal current efficiency of 8.77 cd/A and 10.30 cd/A, respectively. Due to the balanced blue and yellow emission, LPF-TM03 containing 0.3% guest get the relatively pure white light with CIE coordinates value of (0.31,0.33). The results indicate that diarylmaleimide is a kind of utility guest to construct easily synthesized, structurally diverse and highly efficient white polymers.

1. Introduction

White light-emitting materials have attracted increasing interest in both the academia and industry in the last few decades, due to the practical applications in illumination and full-color flat-panel displays [1–5]. An ideal white emission should cover the whole visible range of ca 400–700 nm [6]. However, it is unusual for a single organic compound to emit white light, since the most of luminophores yield luminescence only from the lowest excited state by Kasha's rule. In addition, a single molecule with different fluorophores always produces a dominant emission corresponding to the lowest energy species for energy transfer between fluorophores. Therefore, either two (blue/yellow) or three (blue/green/red) colors emitting materials, including small molecules and polymers, were employed as multiple emissive layers, or mixed together as single emissive layer to realize the white electroluminescence (EL) in the organic light-emitting devices (OLEDs) [7–11].

However, the white emitting systems always encounter the problems of phase separation and bias-dependent electroluminescent spectra, which result in the decrease of device efficiency [8,10]. To

solve the problems, single white polymer systems have been developed by introducing a low-energy fluorophore (guest) into blue-emitting polymer (host) by means of chemical approach [10,12–14]. Through controlling the energy transfer and charge trapping between the guest and host, the highly efficient white electroluminescence with the stable spectra of independent dual-emission was obtained from the white polymer light-emitting devices (WPLEDs) based on the solution-processable single white polymers. Benzothiadiazole fluorophores were often chosen as guest to construct single white polymers for their high photoluminescence and EL efficiencies. For example, benzothiadiazoles were successively introduced into the main chain (M-type) and lateral chain (L-type) of blue-emitting polyfluorene (PF), and the core of star-like (S-type) PF, reaching high EL efficiency of 7–11 cd/A in Ca/Al cathode-based WPLED [12,14–17], and 2.41 cd/A in WPLED with LiF/Al as cathode [18–20]. Due to the intact main chain and branched structure inhibiting intermolecular interactions, L-type and S-type single white PF generally showed higher EL performance than M-type [15,16]. However, because of the difficulty in structure modification of benzothiadiazoles, the complex synthetic procedures were always necessary to covalently link the guest into the different locations of linear

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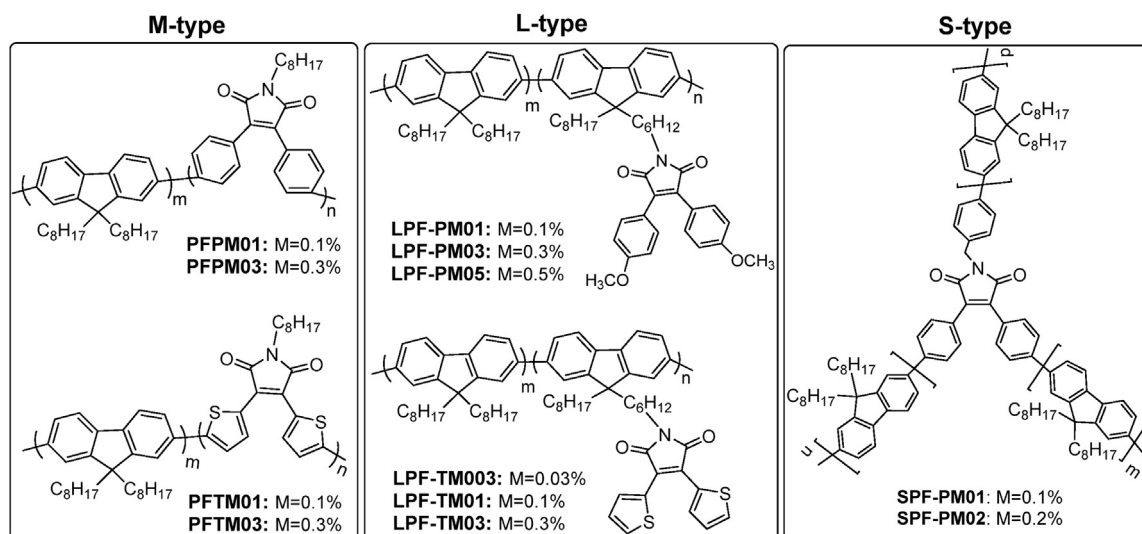


Fig. 1. Structure of single white polymers based on diarylmaleimide (M) guest.

and star-like PF [13,17,20–22].

3,4-Disubstituted arylmaleimide derivatives, one kind of fluorescent dyes first found in natural product [23], have attracted great attentions in the fields of OLED, anion sensing and data storage and process, due to their strong luminescent intensity in solution and solid state [24–31]. Maleimides with three active sites are easy to be branched and substituted by different groups. Due to the electron-deficiency of maleimide, a pair of cross-intercepted dipoles would be formed, when its C(3) and C(4) positions are symmetrically substituted by electron-pushing aryl groups. Thus, the optic band of 3,4-disubstituted arylmaleimide could be adjusted by changing different aryls, realizing different color emission [32]. Just recently, a green diphenylmaleimide and a yellow dithienylmaleimide were introduced as guest into the main chains of linear PF (M-type) to produce single white polymer PFPMs and PFTMs in our laboratory (Fig. 1). PFPMs displayed a purer white emission than PFTMs for the large red-shift in emission of guest in the backbone of PF, originating from the extended conjugation length. PFPM03 exhibited a white EL from a solution-process device with the configuration of ITO/PEDOT:PSS/polymer/TPBI/LiF/Al, reaching a maximum current efficiency of 8.14 cd/A [33]. Under the stimulation of flexible modification of maleimide structure, in this work, we employed di(*p*-methoxyphenyl)maleimide, dithienylmaleimide and diphenylmaleimide as guests, to synthesize L-type polymers (LPF-PMs and LPF-TMs) and S-type polymers (SPF-PMs), which were expected to be excellent luminescent materials in WPLEDs (Fig. 1).

2. Experiments

2.1. Materials and characterization

All the reagents were purchased from commercial suppliers. Toluene was distilled from CaH₂ under nitrogen gas. Compound 1–3, 6–9, 11, 12, MTM and MPM were prepared by using literature methods [29,32–34]. All reactions were carried out under a nitrogen atmosphere and performed using Schlenk techniques.

High-resolution electrospray ionization mass spectrometry was performed on a TSQ Quantum Access MAX of Thermo Fisher Scientific. ¹H and ¹³C nuclear magnetic resonance (NMR) spectra were recorded on a Bruker Ascend 400 (400 MHz for proton, 100 MHz for carbon) spectrometer with tetramethylsilane as the internal reference using CDCl₃ as solvent in all cases. Ultraviolet-visible (UV-vis) spectra were recorded on a Shimadzu UV-2600 Spectrophotometer. The fluorescence spectroscopy (PL) emission spectra were obtained with a Shimadzu RF-5301 PC spectrophotometer. Cyclic voltammetry (CV) was performed

on a CHI600D electro-chemical analyzer in anhydrous THF containing tetra-*n*-butyl-ammonium hexafluorophosphate (TBAPF6, 0.1 M) as supporting electrolyte at 298 K. A conventional three electrode cell was used with a platinum working electrode and a platinum wire as the counter electrode. The Pt working electrode was routinely polished with a polishing alumina suspension and rinsed with acetone before use. The measured potentials were recorded with respect to Ag/AgCl reference electrode. All electrochemical measurements were carried out under an atmospheric pressure of nitrogen. Thermo gravimetric analysis (TGA) was measured by a Mettler 851e with a heating rate of 20 °C min⁻¹ under flowing N₂. Number-average molecular weight (*M_n*) and the polydispersity index (PDI) of the polymer were determined by gel-permeation chromatography (GPC) on a Waters 1515 instrument. Polystyrene and THF were used as the standard and eluent, respectively.

2.2. Theory calculation

Molecular simulations were performed on Gaussian 09 program. Geometry at the ground state was fully optimized by the density functional theory (DFT) method with the Becke three-parameter hybrid exchange and the Lee-Yang-Parr correlation functional (B3LYP) and 6-31G (d) basis set. The optical transitions were analyzed by time-dependent DFT (TD-DFT) using the B3LYP/6-31G (d) basis set

2.3. OLED fabrication

The devices were fabricated by following processes. First, ITO coated glass substrates were cleaned successively using detergent, deionized water, acetone and isopropanol in an ultrasonic bath, and then dried in drying cabinet followed by pretreatment with oxygen plasma. Then the poly-(ethylenedioxythiophene):poly(styrenesulfonic acid) (PEDOT:PSS), and the resulted polymers were deposited by the spin coating. Finally, 2,2,2-(1,3,5-benzenetriyl)tris(1-phenyl)-1H-benzimidazol (TPBI), Lithium fluoride (LiF) and Al metal were deposited by the thermal evaporation under a base vacuum of about 3 × 10⁻⁵ Pa. The thickness of the films was determined by quartz crystal monitors. The active area of the EL device, defined by the overlap of the ITO and the cathode electrode, was 2 mm × 2 mm. Current density-voltage, luminance-voltage, current efficiency-current density and power efficiency-current density characteristics were measured with a computer controlled Keithley 2400 Source Meter and a PR-655 Spectrophotometer at ambient atmosphere.

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