



All-solution-processed multilayer polymer/dendrimer light emitting diodes



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ABSTRACT

A fully solution-processed deep-blue emitting organic light emitting diode (OLED) based on a highly efficient fluorescent dendritic material with a pyrene core, a phenylene shell and triphenylamine surface groups coupled with polymeric hole (HTL) and electron (ETL) transport layers is demonstrated. Each layer formed smooth and pinhole-free films as demonstrated by Atomic Force Microscopy (AFM) as well as by X-ray Photoelectron Spectroscopy (XPS). Furthermore, detailed Ultraviolet Photoelectron Spectroscopy (UPS) surveys revealed a beneficial energy level alignment and hence improved charge carrier confinement. The resulting triple-layer device saw a 7.7-fold increase in current efficiency compared to a single-layer device while maintaining a deep-blue emission color characterized by the CIE1931 coordinates of $x = 0.153$ and $y = 0.155$.

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1. Introduction

Ever since the first successful demonstration of a polymer light emitting diode (PLED) [1] the prospects of cheap and large-scale solution-based fabrication led to a significant research interest to advance this technology from lab to production scale. Thus far, however, solution-processed PLEDs often significantly trail the performance achieved by high-vacuum vapour deposition techniques, which diminishes the potential cost savings introduced by a solution process. To increase the attractiveness of solution-processed organic light emitting diodes (OLED), the performance of the employed solution processable organic semiconductors needs to be improved. Often the reduced performance of polymers with respect to small molecules can be directly related to reliable

structure-property relationships [2], which are usually difficult to unravel in polymeric materials [3]. At the same time, a respectable number of small molecular organic semiconductors has been demonstrated to be solution-processable [4–6]. However, compared to higher molecular weight material such as polymers, the small-molecular compounds are often inferior with respect to film-forming capabilities in a solution-based process as well as are prone to recrystallization as a consequence of a low glass transition temperature [7–9]. A combination of the advantages of low and high molecular weight semiconductors can be realized by dendrimers, which are dendritic macromolecules synthesized generation by generation in a highly controlled way [10]. In contrast to polymeric systems with dendritic side-chains [11], this approach allows for a controlled synthesis, perfect site definition and thereby highly reliable structure-property relations [12]. Utilization of appropriate charge transport groups in the dendrimer further ensures balanced charge carrier injection and transport. Moreover, confinement of the exciton to the emissive layer is at least equally important [13,14] so that OLEDs often incorporate several layers where each layer fulfils specific purposes, such as carrier injection,

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transport, electron or hole blocking, and the aforementioned confinement of the exciton in a double-heterostructure device configuration. For OLEDs fabricated by high-vacuum deposition techniques, the realization of any multilayer configuration is easily achievable. Unfortunately, quite the opposite is true in the case of a solution-based process, where great care has to be taken with respect to an erosion of the already deposited layer by the solvents of the succeeding ones. For this purpose crosslinking of the deposited layers [9,15], orthogonal solvents [16,17], thermal stabilization [18,19] and liquid buffer layers [20] have been successfully utilized for the fabrication of solution-processed multilayer devices. While most of these approaches were reported for polymers and small-molecules, the class of dendritic conjugated materials has so far been neglected. Therefore, the realization of fully solution-processed multilayer dendrimer light emitting diodes has so far been hamstrung by the lack of suitable solution-processable hole- and electron-transporting dendrimers. An alternative is provided by the usage of charge carrier transporting/injecting polymers. Consequently, in this contribution, we report on the combination of a solution-processable hole transport polymer Poly [(9,9-dioctylfluorenyl-2,7-diyl)-co-(4,4'-(N-(4-sec-butylphenyl)diphenylamine))] (TFB) as well as an electron transporting polymer 9,9-Bis(3-(5',6'-bis(4-(polyethylene glycol)phenyl)-[1,1':4,1''-terphenyl]-2'-yl)propyl)-9',9'-dioctyl-2,7-polyfluorene (PEGPF) with a highly efficient light emitting dendrimer, PYGTGA, which consists of a pyrene core, a shell with triphenylene units in the first and phenylenes in the second generation with triphenylamine (TPA) units on the surface. The effectiveness of the combination of TFB and PEGPF with PYGTGA was investigated by layer-by-layer atomic force microscopy (AFM), photoelectron spectroscopy measurements as well as by a comparison of single-, bi- and triple-layer devices.

2. Material and methods

The multifunctional emissive dendritic material PYGTGA was synthesized as reported elsewhere [12]. TFB was purchased from Luminescence Technology Corp. (LUMTEC) and used without further purification. PEGPF was synthesized as described elsewhere [21]. The structures of all compounds are depicted in Fig. 1.

X-ray and ultraviolet photoelectron spectroscopy (XPS, UPS) measurements were performed at the end station SurICat

(beamline PM4) of the synchrotron light source BESSY II (Berlin, Germany). Spectra were collected with a hemispherical electron energy analyser (Scienta SES 100). The excitation photon energy was 35 eV for UPS and 620 eV for XPS, the energy resolution was set to 150 meV. Secondary electron cut-off (SECO) spectra to determine the sample work function were recorded while the sample was biased at -10 V to clear the analyser work function. To assure the absence of sample charging, standard procedures, including variation of the excitation photon flux and sample illumination with visible light, were applied for every sample. An error of less than ± 0.1 eV was estimated for reported energy values.

For the fabrication of single- and multilayer-devices all indium tin oxide (ITO) covered glass substrates were cleaned mechanically in acetone and isopropanol. The substrates were subjected to subsequent supersonic cleaning steps in deionized water, toluene and isopropanol. To enhance the wettability of the first polymeric layer, poly(3,4-ethylenedioxythiophene)-polystyrenesulfonic acid (Baytron P VPAI 4083) (PEDOT:PSS) from H. C. Starck, the ITO substrates were subdued to a dry cleaning procedure in oxygen plasma. PEDOT:PSS was spin-cast onto the prepared substrates under ambient conditions and dried according to the specifications under dynamic vacuum with a pressure less than 1×10^{-5} mbar. For single layer OLEDs, PYGTGA was spin cast from an 8 mg ml^{-1} toluene solution. The thicknesses of the PYGTGA layers were determined to 50 nm with a mean error of less than 10%. To fabricate bilayer- and triple-layer-PLEDs, TFB layers were deposited by spin-coating from a 15 mg ml^{-1} solution under inert conditions and dried in argon at 200°C for 2 h. Afterwards, the substrates were cooled to room temperature on a cold surface. To remove the soluble parts of the annealed polymer the TFB layer was spin-rinsed by pure toluene resulting in a final layer thickness of 5 nm with good reproducibility as long as annealing temperature and time were kept constant. Following that, PYGTGA layers were spin-cast onto the prepared TFB films from an 8 mg ml^{-1} toluene solution and dried at 80°C for 2 h in vacuum ($p \leq 1 \times 10^{-5}$ mbar) resulting in layer thicknesses of 75 nm. Prior to depositing PEGPF in the case of triple-layer devices, the TFB/PYGTGA-substrates were spin-rinsed by pure methanol. Afterwards, PEGPF dissolved in methanol was applied by spin-coating at a polymer content of 1 mg ml^{-1} . The layer was annealed at 70°C for 1 h under high vacuum ($p \leq 1 \times 10^{-5}$ mbar) and exhibited a thickness of 10 nm, which was well controllable for spinning speeds above 1500 rpm. After

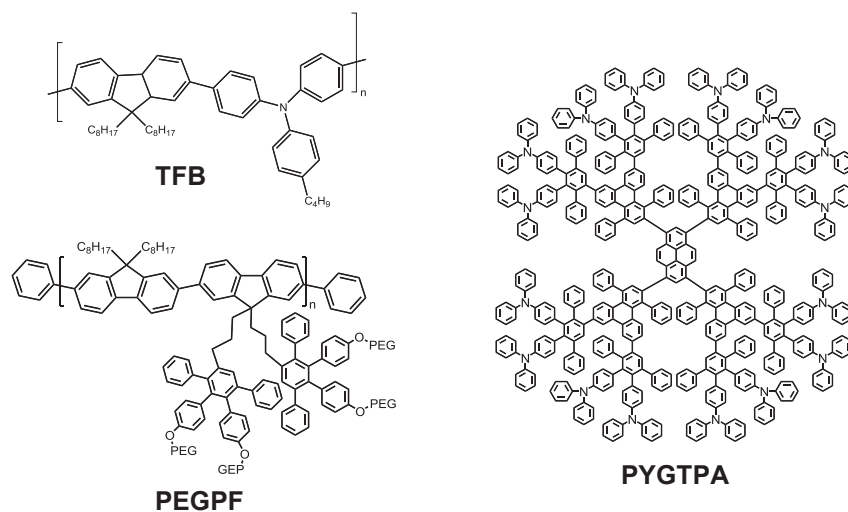


Fig. 1. Chemical structure of the polymers and the dendrimer employed in the solution-processed polymer/dendrimer LED assembly: TFB (top left), PYGTGA (right) and PEGPF (bottom left).

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