



New environmentally friendly polyazomethines with thiophene rings for polymer solar cells

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Received 18 August 2014; received in revised form 27 March 2015; accepted 30 March 2015

Available online 19 May 2015

Communicated by: Associate Editor Hari Mohan Upadhyaya

Abstract

We synthesized two new, air-stable and environmentally friendly polyazomethines and characterized their photovoltaic and electrochemical impedance spectroscopic properties. Two polyazomethines with one (25Th-DMB) or three (2252Th-DMB) thiophene rings were synthesized and tested as donors in the active layer in solar cells. The investigated polymer solar cells contained different amounts of PCBM and PC₇₁BM. The bulk heterojunction polymer solar cells were fabricated and studied, considering the architecture of the devices (i.e., LiF, TiO₂, and TiO₂:Ag) and the fabrication conditions (i.e., air, inert gas, and annealing). The highest power conversion efficiency, which was equal to 0.52% under simulated 100 mW/cm² AM 1.5G irradiation, was determined for the device using polyazomethine with three thiophene rings and PC₇₁BM created in air with the ITO/PEDOT:PSS (100 cP)/2252Th-DMB:PC₇₁BM (1:2)/Al architecture. In addition, the power conversion efficiency was strongly dependent on the viscosity of the PEDOT:PSS used.

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Keywords: Polyazomethines; Thiophene rings; Polymeric photovoltaics; Bulk heterojunction solar cells

1. Introduction

Polyazomethines (PAZs), which is the condensation product of diamine and dialdehyde, are interesting for application as organic photovoltaics because they are inexpensive and require facile purification because water is the

only co-product of the reaction (Iwan and Sek, 2008, 2011). Therefore, PAZ is environmentally friendly and can be applied to generate green energy.

Much effort has been focused on the synthesis of new polymers and small molecule chemical compounds for application in organic solar cells e.g., (Bundgaard and Krebs, 2007; Günes et al., 2007; Zhan and Zhu, 2010; Jayawardena et al., 2013; Cai et al., 2010; Li, 2012; Kesters et al., 2014; Chen et al., 2009; Venkataraman

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et al., 2010; Chen and Hsu, 2011; Pivrikas et al., 2011; Aslan et al., 2014; Scharber and Sariciftci, 2013; Notarianni et al., 2014; Zhou et al., 2012). Commercially available polymers, such as poly(3-hexylthiophene) (P3HT), and poly[N-9"-heptadecanyl-2,7-carbazole-alt-5,5-(4',7'-di-2-thienyl-2',1',3'-benzothiadiazole)] (PCDTBT), have been investigated as donors. Commercially available fullerene derivatives, such as [6,6]-phenyl-C₆₁-butyric acid methyl ester (PCBM) and [6,6]-phenyl C₇₁-butyric acid methyl ester (PC₇₁BM), have been primarily investigated as acceptors [e.g., (Bundgaard and Krebs, 2007; Günes et al., 2007; Zhan and Zhu, 2010; Jayawardena et al., 2013; Cai et al., 2010; Li, 2012; Kesters et al., 2014; Chen et al., 2009; Venkataraman et al., 2010; Chen and Hsu, 2011; Pivrikas et al., 2011; Aslan et al., 2014; Scharber and Sariciftci, 2013; Notarianni et al., 2014; Zhou et al., 2012). Instead of P3HT and PCDTBT, other polymers and copolymers with thiophene, vinylene, benzothiadiazole and carbazole units have been proposed as components for use in polymer solar cells [e.g., (Brabec et al., 2002; Blouin and Leclerc, 2008; Son et al., 2014a,b).

Currently, some azomethines have also been investigated for use as components of the active layer in organic solar cells [e.g., (Jeevadasan et al., 2014; Iwan et al., 2014a,b,c; Moussalem et al., 2014; Canli et al., 2013). For photovoltaic applications, some researchers have propose the use of polyazomethines as the donor in the active layer (Sharma et al., 1996; Hindson et al., 2010; Iwan et al., 2012, 2013a,b, 2011, 2014d; Palewicz et al., 2011). The first study of polyazomethines for solar cells was published by Sharma et al. (1996). The authors studied the photovoltaic properties of a poly(phenyl azomethine furane) (PPAF) cell with an Ag/PPAF/ITO architecture, which had a power conversion efficiency (PCE) of approximately 0.019%. Next, Hindson et al. (2010) analyzed the photovoltaic properties of triphenylamine-based polyazomethines obtained from 2,5-thiophenedicarboxaldehyde, terephthalaldehyde or 1,3-isophthalaldehyde and 4,4'-diaminotriphenylamine, and a PCE of 0.12% was observed.

In our previous studies, we investigated various polyazomethines to determine their suitability for application in organic solar cells (Iwan et al., 2012, 2013a,b, 2011, 2014d; Palewicz et al., 2011). For polyazomethine obtained from 2,7-diaminofluorene and 2,5-thiophenedicarboxaldehyde blended with PCBM, we received a PCE equal to 0.31% was obtained (Iwan et al., 2012). However, other polyazomethine with thiophene and *cardo* moieties synthesized by our group did not exhibit photovoltaic properties (Iwan et al., 2013a). In addition, we studied the photovoltaic properties of liquid crystalline poly(siloxane-azomethines). The obtained PCE was approximately 0.01% (Iwan et al., 2011; Palewicz et al., 2011). Finally, we investigated polyazomethines with a triphenylamine group (Iwan et al., 2013b, 2014d), and a record PCE value of 0.56% was obtained for air-stable aromatic polyazomethine with triphenylamine moieties (Iwan et al., 2014d). In Table 1, the chemical structures and PCEs of all of the investigated

polyazomethines for photovoltaic applications are summarized.

In this study, new polyazomethines with thiophene rings were synthesized via a one-step high temperature condensation technique. We increased the conjugated length of the electron donor (D) moiety by introducing three thiophene rings into the mer unit of 2252Th-DMB. In addition, we introduced methoxy groups to the benzidine moiety to improve the solubility of both synthesized polyazomethines. Polymer solar cells based on the polyazomethines were constructed, and their PEC was investigated based on seven main factors, as follows:

- (i) influence of the number of thiophene rings in the polyazomethine mer units,
- (ii) influence of the type of fullerene derivative (i.e., PCBM and PC₇₁BM),
- (iii) presence of LiF,
- (iv) viscosity of applied PEDOT:PSS (5–12 cP, 100 cP),
- (v) presence of 1,8-dibromooctane (DBO) in the active layer,
- (vi) presence of TiO₂ and the amount of silver in the TiO₂ layer, and
- (vii) type of atmosphere used during the construction of the photovoltaic devices (i.e., air and inert gas).

2. Experimental section

2.1. Materials and synthesis procedure

2,2':5',2''-Terthiophene-5,5''-dicarboxaldehyde, 2,5-thiophenedicarboxaldehyde, 3,3'-dimethoxybenzidine, *N,N*-dimethylacetamide (DMA), chloroform, [6,6]-phenyl C₆₁ butyric acid methyl ester (PCBM), PC₇₁BM, 1,8-dibromooctane (DBO), and poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS) with a viscosity of 100 cP were purchased from Sigma–Aldrich and used as received. Methanol and acetone were purchased from POCH and used as received. ITO and PEDOT:PSS (5–12 cP) were purchased from the Ossila Company. The surface resistance of ITO was approximately 20 ohm/square. Titanium (IV) butoxide (TBOT) (99+ %) was purchased from Alfa Aesar. Silver nitrate and ethanol (96%) were purchased from POCH.

2.1.1. Polymers synthesis

The polymers were obtained using a one-step high temperature condensation technique with anhydrous CaSO₄ as the water trap. A single-neck flask with a magnetic stir bar was charged with dialdehyde (1 mmol), diamine (1 mmol), *p*-toluenesulfonic acid (PTS) and 10 mL of DMA.

The reaction mixture was allowed to stir for 22 h at 160 °C in an oil bath. The polymer solution was precipitated in methanol, and the polymer was collected by filtration. The solid was washed with hot methanol and hot acetone. The final polymer was dried overnight at 80 °C.

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