



Conjugated ionic (co)polythiophene-based cathode interlayers for bulk heterojunction organic solar cells

Sanne Govaerts^a, Jurgen Kesters^a, Maxime Defour^b, Bruno Van Mele^b, Huguette Penxten^a, Shova Neupane^c, Frank Uwe Renner^{c,d}, Laurence Lutsen^d, Dirk Vanderzande^{a,d}, Wouter Maes^{a,d,*}

^a UHasselt – Hasselt University, Institute for Materials Research (IMO), Design & Synthesis of Organic Semiconductors (DSOS), Agoralaan, 3590 Diepenbeek, Belgium

^b Vrije Universiteit Brussel (VUB), Physical Chemistry and Polymer Science (FYSC), Pleinlaan 2, 1050 Brussels, Belgium

^c UHasselt – Hasselt University, Institute for Materials Research (IMO), Energy Materials and Interfaces (EMInt), Wetenschapspark 1, 3590 Diepenbeek, Belgium

^d IMOMEC Division, IMEC, Wetenschapspark 1, 3590 Diepenbeek, Belgium

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ABSTRACT

The incorporation of conjugated polyelectrolytes as cathode interlayers in organic photovoltaics has been proven to be an effective way to boost the device efficiency. Nevertheless, more detailed investigations of the structure–property relationships of these interlayer materials, in particular related to the film deposition behavior, can provide further insights into their mode of action. With this aim, a series of ionic (co)polythiophenes is successfully synthesized via Kumada catalyst-transfer condensation polymerization and subsequent introduction of ionic moieties on the polymer side chains. Both the topology (i.e. homopolymers, random and block copolymers) and the amount of ionic groups are systematically varied. The polymers are fully characterized and then applied as cathode interlayers in polymer solar cells based on PCDTBT:PC₇₁BM, affording an average efficiency increase of ~15%. The structural screening on one hand indicates that the efficiency gain is a rather general phenomenon for this material class. On the other hand, the best photovoltaic responses are observed for the conjugated polyelectrolytes with a higher triethylene glycol side chain ratio and the block copolymer structure performs slightly better as compared to the random copolymer with the same (50/50) monomer ratio. Based on these findings, the field can move on to a more rational development of novel interfacial materials and thereby push the device efficiency even further.

1. Introduction

Over the past decade, organic photovoltaics (OPVs) have shown a growing potential as an alternative renewable energy source, in particular because of some additional appealing features such as flexibility, semi-transparency, improved low-light performance, and low-cost (high-throughput) large area production [1–3]. The introduction of the bulk heterojunction (BHJ) concept, in which the photoactive layer consists of electron donating and accepting materials intimately mixed at the nanoscale, the development of push-pull type low bandgap organic semiconductors to enhance the solar spectrum coverage, and the use of optimized device architectures have been key developments to elevate the power conversion efficiency (PCE) of OPV devices to values currently exceeding 12% [4–8]. Even though most research activities have been oriented on the development of novel active layer materials with appropriate optoelectronic properties, the introduction of interlayer materials has become a widely accepted approach to further

enhance the device efficiency [9–17]. One class of interlayer materials of particular interest are conjugated polyelectrolytes (CPEs). They can be processed from eco-friendly, orthogonal solvents, thereby preventing re-dissolution of the underlying layer during device fabrication. The incorporation of such CPEs has led to PCE improvements of more than 20% compared to reference devices, which can mainly be attributed to the formation of interfacial dipoles, influencing the work function of the adjacent electrode [10,18–23]. This improves the energy level alignment between the photoactive layer and the cathode (in standard device stacks), resulting in an enhanced charge collection and a reduced charge recombination.

Several CPE materials have already been studied as cathode interlayers. The conjugated polymer backbone has not been explored much further than polyfluorene and polythiophene derivatives and most variation has been introduced in the ionic moieties [12]. Previous work in our group focused on imidazolium-functionalized polythiophenes, outperforming the ammonium-functionalized counterparts [11]. More

* Corresponding author at: UHasselt – Hasselt University, Institute for Materials Research (IMO), Design & Synthesis of Organic Semiconductors (DSOS), Agoralaan, 3590 Diepenbeek, Belgium.

E-mail address: wouter.maes@uhasselt.be (W. Maes).

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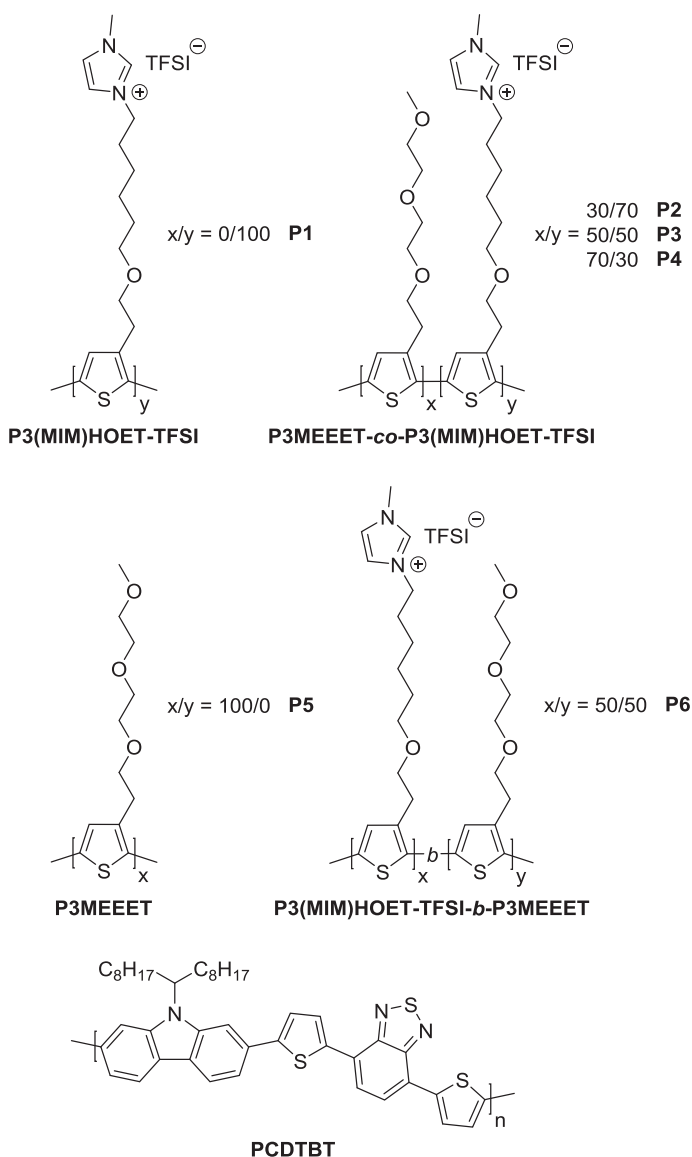


Fig. 1. Overview of the synthesized polymers and the used donor material.

recently, impedance spectroscopy measurements revealed that the dielectric permittivity, induced by the ionic functionalities, is an important parameter to improve charge collection [23]. The exact CPE structure also influences other aspects. An ionic random (50/50) copolythiophene with triethylene glycol and imidazolium-functionalized side chains (**P3**; Fig. 1) for instance demonstrated superior performance in comparison to a fully imidazolium-functionalized homopolymer, resulting from an improved affinity with the photoactive layer beneath due to the presence of the non-ionic side chains [23]. For inverted OPV devices incorporating the CPE at the bottom side, Chen et al. demonstrated that more smooth and homogeneous photoactive layer deposition could be obtained by increasing the amount of polar groups in polyfluorene-based CPEs [22].

In this work, a series of polythiophene CPEs with TFSI (bis(trifluoromethane)sulfonimide) counteranions was synthesized by Kumada catalyst-transfer condensation polymerization (KCTCP), subsequent substitution of the bromide-functionalized side chains, and final exchange of the Br counteranions. Due to the controlled chain-growth character of this type of polymerization, different polymer topologies (homopolymers, random and block copolymers) were obtained with a high control over the molar mass, dispersity and end groups in a relatively straightforward way via one-pot procedures. The ratio of the ionic groups present in the random copolymers was systematically

varied and a block copolymer was prepared as well for comparison. The synthesized materials were then evaluated as cathode interlayer materials in standard BHJ OPV devices based on PCDTBT:PC₇₁BM to further explore the effect of structural changes on the overall device performance.

2. Results and discussion

2.1. Polymer Synthesis

From previous work, the **P3MEEET-co-P3(MIM)HOET-TFSI** (50/50) random copolymer (**P3**; Fig. 1) was identified as the CPE material affording best OPV device efficiencies for various photoactive layer blend combinations [23–25]. This material was hence used as the starting point to create further structural variation. A series of random copolymers with monomer ratios varying from 0/100, 30/70, 50/50, 70/30 to 100/0 was targeted. For the synthesis of these random copolymers, the two different precursor monomers were first prepared, i.e. 2,5-dibromo-3-[2-(2-(2-methoxyethoxy)ethoxy)ethyl]thiophene (**pM1**) and 2,5-dibromo-3-[2-(6-bromohexyloxy)ethyl]thiophene (**pM2**) (Scheme 1) [26]. These precursor monomers were then activated via a Grignard metathesis (GRIM) reaction. This was done prior to the polymerization by combining **pM1** and **pM2** in the respective ratio in

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