

A PCPDTTPD-based narrow bandgap conjugated polyelectrolyte for organic solar cells

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ABSTRACT

Extensive research on organic photovoltaics has granted impressive power conversion efficiencies, nowadays exceeding 13% for state-of-the-art photoactive material combinations. Nevertheless, different strategies can be adopted to further enhance the efficiency and the competitiveness with alternative photovoltaic technologies. Conjugated polyelectrolytes have been applied as anode or cathode interlayers to optimize ohmic contacts and lower the contact resistance, thereby improving the ultimate device efficiency. Here, we present an interlayer material belonging to the emerging class of narrow bandgap conjugated polyelectrolytes, based on an imidazolium functionalized 4*H*-cyclopenta[2,1-*b*:3,4-*b'*]dithiophene (CPDT) as the electron-rich polymer building block and 4*H*-thieno[3,4-*c*]pyrrole-4,6(5*H*)-dione (TPD) as the electron-deficient subunit. The ionic polymer is applied as cathode interlayer for PBDTTPD:[70]PCBM (poly[bis(2'-ethylhexyloxy)benzo[1,2-*b*:4,5-*b'*]dithiophene-*alt*-*N*-octylthieno[3,4-*c*]pyrrole-4,6-dione]:[6,6]-phenyl-C₇₁-butyric acid methyl ester) bulk heterojunction polymer solar cells, improving the overall device performance from 6.9 to 7.8%.

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1. Introduction

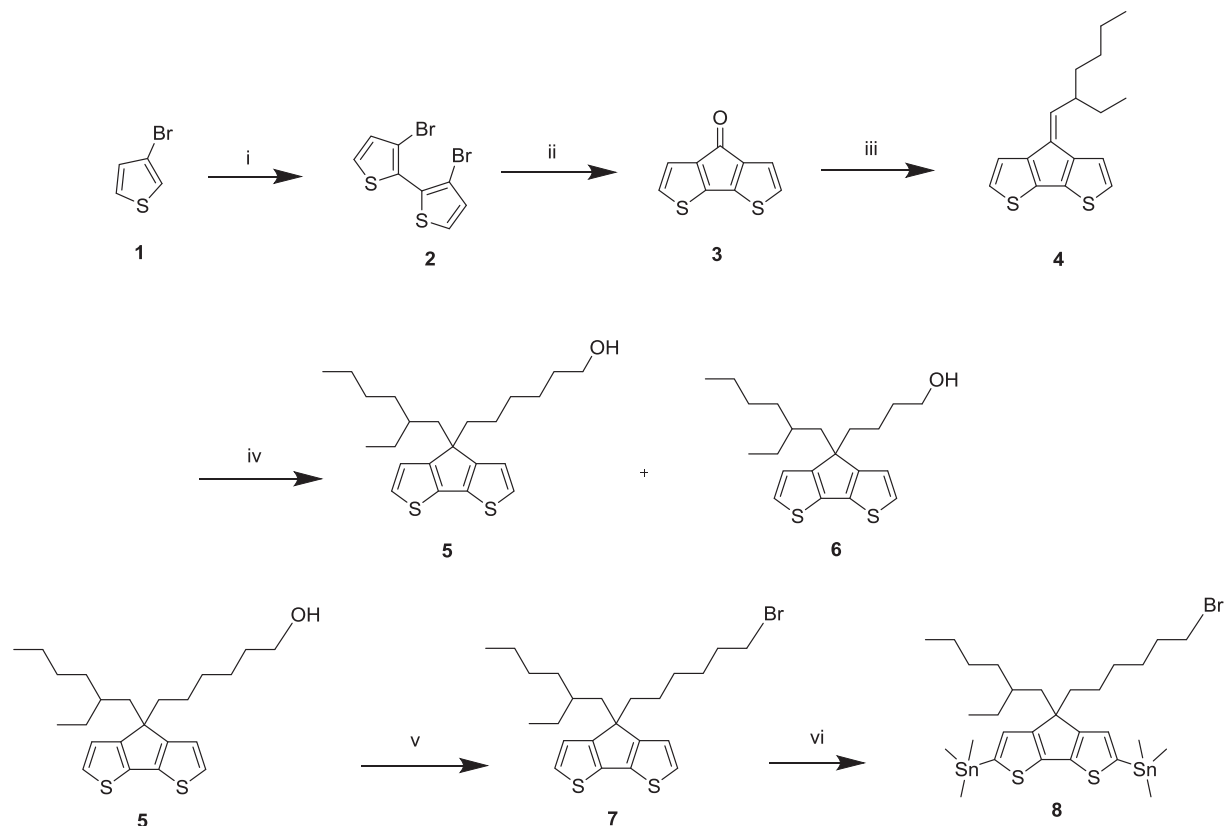
Organic photovoltaics (OPV's) have witnessed a strong growth over the last two decades as a promising technology to convert solar irradiation into electricity [1–4]. In contrast to their silicon-based counterparts, fully flexible and light-weight devices can be targeted via large-area production methods and with a reduced cost [5–8]. On the active organic material side, strong efforts have been directed toward the development of new low bandgap materials (polymers as well as small molecules) that optimally match with the solar spectrum [9,10]. Furthermore, perfect frontier orbital (HOMO and LUMO) energy level alignment of the electron donor and acceptor materials combined in the bulk heterojunction (BHJ) blend and an optimal interpenetrating nanostructured BHJ

morphology are required to maximize the device performance [11,12]. This has resulted in power conversion efficiencies (PCE's) nowadays exceeding 13% for the best donor-acceptor combinations [13–15]. An important point of attention is the efficient charge extraction of the created excitons upon light absorption because of the inherent recombination processes present in (blends of) organic materials [16,17]. BHJ OPV devices typically consist of different layers stacked on top of each other (vide infra). Interface engineering is hence of utmost importance to realize ohmic contacts (with low contact resistance) of the photoactive layer with the metal electrodes and to improve the selectivity of charge transport [18]. Recent studies in this direction have demonstrated the high potential of interface engineering to reduce charge accumulation and to increase charge extraction, enhancing the device performance [19–21].

Conjugated polyelectrolytes (CPE's), combining a conjugated polymer backbone with ionic (mostly side chain) groups, have already proven to be successful as cathode and anode modification layers to boost the photovoltaic performance of OPV and hybrid

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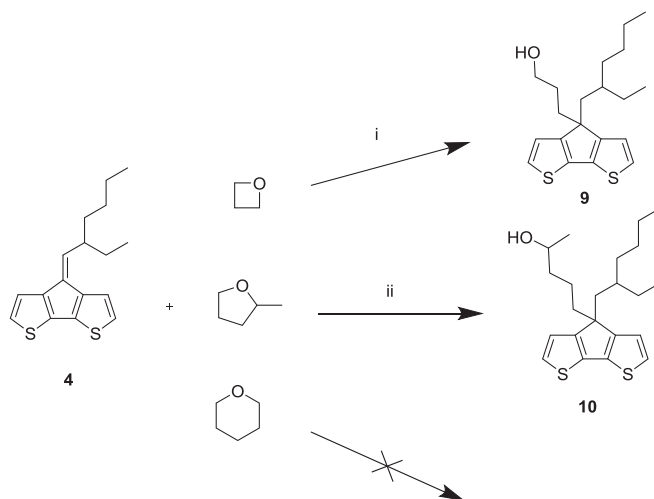


Scheme 1. CPDT monomer synthesis: i) LDA, CuCl₂, THF, overnight at RT (72%); ii) *n*-BuLi, ClCONMe₂, THF, overnight at RT (76%); iii) 2-ethylhexylphosphonium bromide, *n*-BuLi, THF, 2 h at –78 °C, overnight at RT (77%); iv) 1. (6-bromohexyloxy)triisopropylsilane, LiAlH₄, MTBE, overnight at 0 °C; 2. TBAF (71% over two steps); v) PPh₃, CBr₄, 3 h at RT (94%); vi) *n*-BuLi, SnMe₃Cl, 1.5 h at –78 °C (54% after prep-SEC).

organic-inorganic (perovskite) devices [22–25]. Furthermore, the pending polar groups give rise to additional interesting features. An inherent benefit of creating more hydrophilic polymers is their processability from more environmentally friendly, low-boiling solvents (e.g. alcohols) [23,26]. The detailed working principle of CPE interlayers is, however, not always fully understood, in particular with respect to the structural features giving rise to the observed improvements of the different photovoltaic output parameters. So et al. investigated the effect of different interlayers for diverse systems and summarized the different roles these interlayers have [17]. They found that the interlayers control i) the electrode-polymer energy alignment, ii) the built-in electric field, iii) the surface energy, and iv) the surface recombination. Furthermore, interlayers are also applied to prevent penetration of the thermally evaporated electric contact (e.g. Al) into the organic layer. Most importantly, CPE's serve as a simple and powerful tool to enhance the OPV device parameters [22]. The open-circuit voltage (V_{oc}) often improves, mainly because of a higher built-in potential created by the hydrophilic surface and better dipole alignment. The fill factor (FF) also increases due to the ohmic contact with more balanced charge injection and better charge carrier transportation and collection at the respective electrode, hence also improving the short-circuit current density (J_{sc}) [18–20,22,27].

For the design of cathode interlayers, it is preferable to have a low work function and good (thermal) stability. Most CPE's are derived from polythiophenes (often functionalized P3HT derivatives) or polyfluorenes (e.g. PFN) [28]. Although PFN is the most commonly used CPE interlayer material, polythiophenes have recently shown slightly better device performances [21–23,29–31]. A drawback of these CPE's is their tendency to not

uniformly distribute on top of the active layer when spin-coated, leading to the formation of clusters [21,23]. Narrow bandgap conjugated polyelectrolytes (NBGCPE's) are an emerging class of CPE's, aiming to combine the advantages of conjugated low bandgap polymers (e.g. improved compatibility [32], conductivity [33] and charge carrier mobilities [34,35]) and CPE interlayer materials. In 2013, the group of Bazan was the first to demonstrate that NBGCPE's can exhibit superior characteristics (e.g. charge transport



Scheme 2. Reactions of precursor 4 with cyclic ethers: i) MTBE, LiAlH₄, overnight at 60 °C (65%); ii) LiAlH₄, overnight at 75 °C (44%).

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