



A readily-accessible, random perylene diimide copolymer acceptor for all-polymer solar cells



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ABSTRACT

A new, structurally random perylene diimide (PDI) copolymer acceptor, **PPDI**, has been readily synthesized from Stille cross-coupling polycondensation of two pairs of regioisomeric dibromo-PDIs with 2,5-bis(trimethylstannyl)thiophene. **PPDI** possesses a complex structural configuration with a greatly twisted conformation, and thus exhibits weak intermolecular interactions. A solution of **PPDI** and a polymer donor such as PBT7-Th affords a smooth homogenous film without large crystalline domain formation. This composite film absorbs strongly throughout the visible spectrum, and when used as an active layer in all-polymer solar cells, provides a power conversion efficiency of over 5%.

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1. Introduction

Over the past two decades, great progress has been made in solution-processed bulk heterojunction (BHJ) polymer solar cell (PSC) research [1–3]. Many of these BHJ-PSCs have utilized an active layer composed of a composite film of a polymer donor and a fullerene derivative as an acceptor. Due to fullerene's propensity to form relatively large aggregates in the active layer over time with ensuing degraded photovoltaic performance [4], a significant effort has in recent years been devoted to other organic acceptor materials such as imide and diimide derivatives as fullerene replacements [5–7]. In addition, utilization of active layers composed of a polymer acceptor and a polymer donor blends as is generally referred to as all-polymer solar cells (all-PSCs) [6,7] has also become prevalent. This may be attributed to the ease with which these materials can be readily processed by solution techniques and that the active layers of compatible polymer blends may exhibit better thermal stability than those using a polymer or small

molecular donor with a fullerene derivative [4,6,7]. While many imide or diimide polymers have exhibited good electron transport properties [8] and have been employed as acceptors in all-PSCs [9–17]. Although many perylene diimide (PDI)-based polymers have been employed as acceptors in all-PSCs in recent years [6,7,18–27], there are still relatively few PDI polymer acceptors show power conversion efficiencies (PCEs) over 5%.

The first all-PSCs employing a PDI polymer acceptor was reported in 2007 with a power conversion efficiency (PCE) of slightly over 1% [28]. Since then, progress has been made in the PDI polymer acceptor design with the current PCE reaching about 7.5% [14]. This is particularly puzzling in view of the fact that small molecular PDI derivatives have been shown to afford commendably PCEs of over 9% [29,30]. Much like small molecular PDI acceptors [31–33], the performance of PDI polymer acceptors hinges critically on controlling their aggregation behaviors to impede excessive aggregation which would disrupt charge transport pathways in the active layer. For example, 3-dimensional structural framework has been engaged in small molecular PDI acceptor design with the objective of restraining aggregation to the nanoscale levels. This design strategy has met with varying degrees of success, with the most noteworthy one being the spiro difluorene-bi-PDI acceptor

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attaining a PCE of 9.5% [29]. However, for polymer PDI acceptors, the 3-dimensional structural framework design to modulate aggregation behavior may not be as feasible since this may lead to excessive detrimental suppression as some degrees of aggregation may be necessary for efficient charge transport. We believe that a random polymer structure comprising regioisomeric PDI moieties may be utilized in controlling the PDI aggregation of a polymer acceptor.

We report herein the synthesis and electrical and photovoltaic performance of a new, structurally random PDI copolymer acceptor, **PPDI**, comprising essentially four thienyl-derivatized PDI structures differing only in *N*-alkyl-substitutions or/and polymerization sites (Scheme 1). The structural complexity of **PPDI** was to be expected since two different *N,N'*-bis(alkyl)-substituted PDIs, each with two different dibromo substitution patterns, were utilized in the Stille cross-coupling polycondensation with 2,5-bis(trimethylstannyl)thiophene. The engagement of two different *N,N'*-bis(alkyl)-substituted PDIs was intended to enhance the solution processability of the final polymer product as our earlier preparation using one single *N,N'*-bis(2-ethylhexyl)PDI (**PDI-1**) (see Scheme 1) led to insoluble product. In addition, bromination of each *N,N'*-bis(alkyl)-substituted PDI readily afforded two isomeric

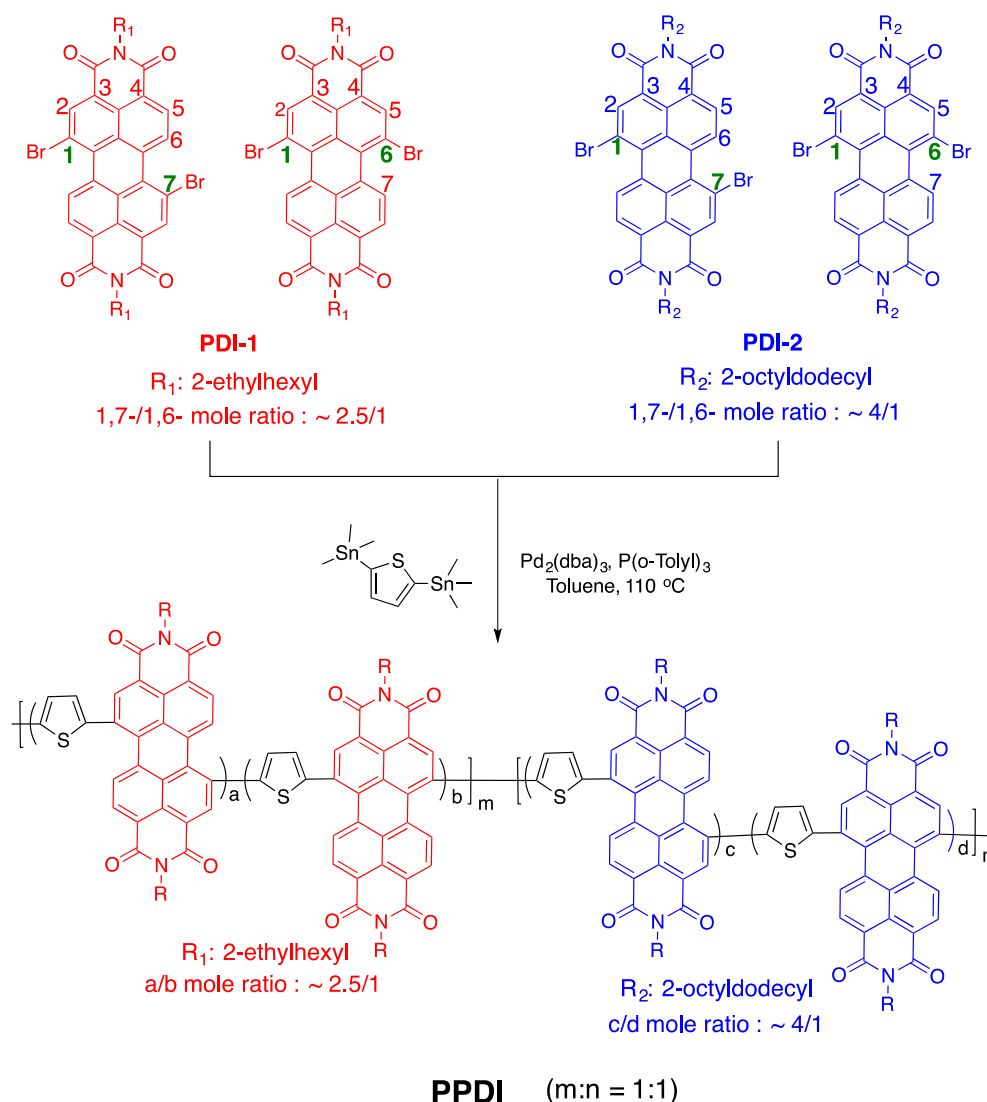
dibromides (i.e. 1,6-dibromo and 1,7-dibromo products) [34] in high yields, which were directly utilized in subsequent polymerization without separation. Accordingly, the resulting structurally random copolymer, **PPDI**, would be expected to provide excellent solution processability characteristics owing to its structural randomness as well as exhibit sterically twisted conformation that discouraged excessive PDI aggregation.

2. Experimental section

2.1. Materials and instruments

N,N'-bis(2-ethylhexyl)-(1,7&1,6)-dibromo-3,4,9,10-perylenetetracarboxylic diimide was purchased from Derthon optoelectronic materials science technology Co LTD. *N,N'*-bis(2-octyldodecyl)-(1,7&1,6)-dibromo-3,4,9,10-perylenetetracarboxylic diimide was prepared according to reported procedure [35]. Other chemicals and reagents were purchased from Aldrich and used without further purification. All the reactions were carried out under nitrogen atmosphere.

Nuclear magnetic resonance (NMR) spectra were conducted on a Bruker Ultrashield 400 Plus NMR spectrometer. Gel permeation



Scheme 1. Synthesis of **PPDI** copolymer acceptor.

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