



# Effect of compositions of acceptor polymers on dark current and photocurrent of all-polymer bulk-heterojunction photodetectors

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## ABSTRACT

A series of electron-acceptor polymers, copolymers and blends were used in all-polymer BHJ photodetectors and the effect of acceptor compositions on the key device parameters of dark current density and photocurrent was investigated. Compared with acceptor polymers and polymer blends, the devices based on acceptor copolymer showed lowered dark current and higher photocurrent, due to optimal molecular stacking and morphology of the BHJ active layer. The acceptor blends tend to cause a large phase separation and rough surface of the active layer, thus leading to a low detectivity of the device. Among all the acceptor compositions studied in this work, the all-polymer BHJ photodetector based on a donor polymer (**PolyD**) and an acceptor copolymer (**PolyAA'50**) exhibited the highest specific detectivity of over  $10^{12}$  Jones in the spectral region of 320–980 nm under  $-0.1$  V bias.

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## 1. Introduction

Dark current and photocurrent of a bulk-heterojunction (BHJ) polymer photodetector (PPD) are key parameters that relate to the device properties (e.g., external quantum efficiency or EQE and spectral responsivity) and figure-of-the-merit (e.g., specific detectivity) [1–5]. Typically, if the dark current is a dominant contributor to the electronic noise, the high dark current at negative bias would decrease the signal/noise ratio, thus weakening the detectivity of the PPDs [6]. On the other hand, the high photocurrent is desirable in order to increase the sensitivity of the PPDs in the ultraviolet (UV), visible (vis), and near-infrared (NIR) spectral regions [7,8]. Similar to organic BHJ solar cells, high charge-transport mobility and well-matched energy levels of the donor and acceptor promote

charge separation and transport, thus affording high EQE and high photocurrent [9,10]. Furthermore, morphology of the BHJ layer can greatly affect the dark current, which in turn relates to the types and compositions of polymer donors and acceptors [11,12].

To improve the performance of PPDs, various strategies of lowering the dark current and enhancing the EQE and photocurrent have been explored [13–15]. Recently, the ternary polymer blends consisting of two electron acceptors and one donor were used to form a BHJ active layer, which opens up an opportunity of exploring the multi-component BHJ all-polymer PPDs [16–18]. For example, Bo and co-workers showed that a BHJ ternary system by using a non-fullerene acceptor together with PC<sub>71</sub>BM acceptor and polymer donor could lead to the improved EQE and higher photocurrent [19]. On the other hand, copolymers comprising the two different acceptor or donor segments could be considered to be an alternative to multi-component polymer blends for use in all-polymer PPDs [20–22]. For example, our group recently demonstrated that the morphology of the BHJ layer and device performance are dependent of the ratios of the different donor (A<sub>1</sub> and A<sub>2</sub>) segments in a series of random A<sub>1</sub>-D-A<sub>2</sub>-D copolymers [23].

Although the multi-component BHJ system can be readily made

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available by either blending the different acceptor and donor polymers or by using acceptor or donor copolymers, it is difficult to predict the effect of acceptor compositions on device performance [24,25]. Unfortunately, such a comparative study is still lacking, especially with regard to direct comparison of acceptor polymer blends with acceptor copolymers in all-polymer photodetectors. Herein, we report the comparative study of the effect of the blend of two acceptor polymers and structurally related acceptor copolymers on the dark current and photocurrent of all-polymer photodetectors. Our study revealed that the compositions of polymer acceptors, either as a blend or copolymer, affect the absorption profile and film morphology of the BHJ layer, which in turn affects the device performance and the use of acceptor copolymer with the same donor polymer is more effective than the related acceptor blend for lowering the dark current and enhancing the photocurrent of all-polymer photodetectors.

## 2. Results and discussion

### 2.1. Synthesis and characterization of donor and acceptor polymers

A known donor polymer (**PolyD**) and the two known acceptor polymers **PolyA** and **PolyA'** were chosen in our study (Fig. 1) [26–28]. These acceptor polymers share a common moiety of *N,N'*-bis(2-octyldodecyl)-2,6-dibromonaphthalene-1,4,5,8-bis(dicarboximide) (NDI) and differ in the linkage (thiophene and dithiophene). The acceptor copolymers (**PolyAA'**) are designed to have the same NDI, thiophene and bithiophene units in different ratios, for comparison with the blends of **PolyA** and **PolyA'** in different ratios (Fig. 1). Three acceptor copolymers were synthesized from the corresponding three known monomers in different ratios [29–31], with the **PolyA'** segment of 25% (**PolyAA'25**), 50% (**PolyAA'50**) and 75% (**PolyAA'75**), respectively. For comparative study, the acceptor blends **PolyA/A'** were prepared from **PolyA** and

**PolyA'** in 3:1, 1:1 and 1:3 wt ratios, denoted as **PolyA/A'25**, **PolyA/A'50** and **PolyA/A'75**, respectively. These acceptor blends are deemed to be compositional equivalents to copolymers **PolyAA'25**, **PolyAA'50** and **PolyAA'75**.

The copolymers are readily soluble in common organic solvents, such as tetrahydrofuran (THF), chloroform and chlorobenzene. The chemical structures of the copolymers were confirmed by <sup>1</sup>H NMR spectroscopy and element analysis. The molecular weights were measured by gel permeation chromatography (GPC) using polystyrene as standards in chloroform at room temperature. The number-averaged molecular weights (*M<sub>n</sub>*) of **PolyA**, **PolyAA'25**, **PolyAA'50** and **PolyAA'75** and **PolyA'** were measured to be 203.9, 41.0, 32.8, 30.9 and 23.6 kg/mol, respectively (Table 1).

### 2.2. Thermal properties

Thermogravimetric analysis (TGA) and differential scanning

**Table 1**  
Characterizations of acceptor polymers and copolymers.

	<i>M<sub>n</sub></i> (kDa) <sup>a</sup>	PDI <sup>a</sup>	<i>T<sub>d</sub></i> (°C) <sup>b</sup>	$\lambda_{\text{max}}^{\text{soln.}}$ (nm) <sup>c</sup>	$\lambda_{\text{max}}^{\text{film}}$ (nm) <sup>d</sup>	<i>E<sub>g</sub></i> <sup>opt.</sup> (eV) <sup>e</sup>	HOMO (eV) <sup>f</sup>	LUMO (eV) <sup>f</sup>	<i>E<sub>g</sub></i> <sup>EC</sup> (eV) <sup>g</sup>
<b>PolyA</b>	203.9	1.5	450	708	701	1.38	−5.93	−3.97	1.96
<b>PolyAA'25</b>	41.0	2.7	450	618	666	1.48	−5.93	−3.95	1.98
<b>PolyAA'50</b>	32.8	2.1	450	577	629	1.59	−5.93	−3.86	2.07
<b>PolyAA'75</b>	30.9	2.0	450	555	606	1.60	−5.93	−3.86	2.07
<b>PolyA'</b>	23.6	1.8	450	544	600	1.75	−5.93	−3.85	2.08

<sup>a</sup> Measured by GPC with polystyrene standards in chloroform at room temperature.

<sup>b</sup> Onset temperature for 5% weight loss in nitrogen by TGA.

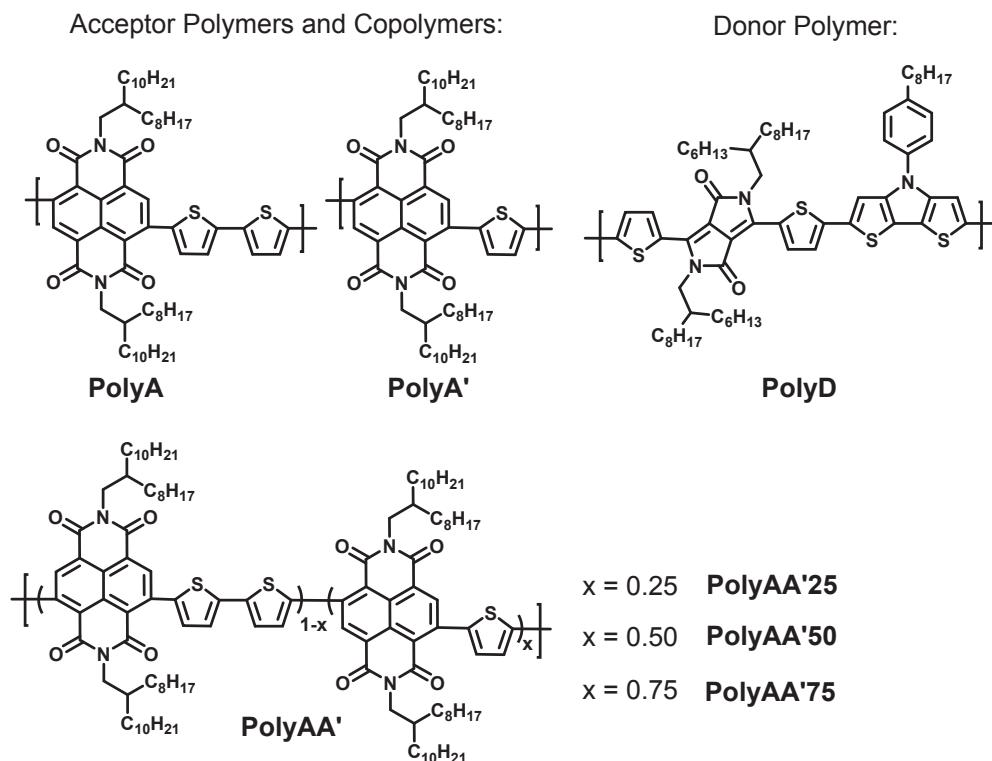
<sup>c</sup> Measured in chloroform at a concentration of 0.03 g/L.

<sup>d</sup> Film spin-cast from 10 mg/mL chloroform solution on quartz substrate.

<sup>e</sup> Optical band gap.

<sup>f</sup> Calculated from  $E_{\text{LUMO}} = -e(E_{\text{on}}^{\text{red}} + 4.43)$  and  $E_{\text{HOMO}} = -e(E_{\text{on}}^{\text{ox}} + 4.43)$ .

<sup>g</sup> Electrochemical band gap is derived from  $E_{\text{HOMO}}$  subtracting  $E_{\text{LUMO}}$ .



**Fig. 1.** Donor polymer and acceptor polymers and copolymers used in all-polymer BHJ photodetectors.

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