

## Induced photodegradation of quinoxaline based copolymers for photovoltaic applications

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### ABSTRACT

We report here the synthesis and characterization of a series of p-type copolymers, which combine a fluorinated quinoxaline (FQ) acceptor unit either with a differently substituted benzodithiophene (BDT) or an unsubstituted thieno[3,2-b]thiophene (TT). The effect of the structural modifications on the photochemical stability of the resulting films is investigated and then correlated with the photovoltaic performance and lifetime measurements of corresponding photovoltaic devices. To this end, we firstly studied the intrinsic stability of each polymer film by monitoring the UV–vis absorption decay, under simulated sunlight, as a function of ageing time. Bulk heterojunction solar cells, based on these polymers as donor materials, were fabricated and tested. Beside the initial values, we monitored the photovoltaic performance during prolonged light soaking in order to evaluate and compare the photostability of more complex systems such as working solar cells.

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### 1. Introduction

Organic Photovoltaics (OPVs) represent one of the latest emerging technologies in solar power generation that offers, with its unique properties, the opportunity to fulfill specific requirements in application areas that have been traditionally difficult for the current PV technologies. The main advantages of OPV devices are the low energy consuming, low cost manufacturing processes, large area coverage and modulation of esthetics properties such as color, transparency, shape and design, in combination with flexible and light-weight substrates [1].

Recently, the performance of laboratory-scale bulk heterojunction (BHJ) polymer solar cells has been greatly improved reaching power conversion efficiencies (PCEs) over 10% and 11% respectively for single [2]- and multi [3]-junction devices, through the synergic development of novel materials [4], enhanced understanding of polymeric

film microstructure [5] and meticulous device optimization [6], indicating a bright future for innovative commercial applications [7].

However, despite the great advances of OPV cells in terms of efficiency and processing, a big concern is still represented by the lifetime of the resulting devices [8]. The performance decay of BHJ solar cells can be mainly ascribed to several degradation mechanisms occurring at various rates and regions of the device. The main triggers of degradation have been clearly identified: oxygen, moisture, temperature and light. For instance, diffusion of water and oxygen into the device promotes chemical degradation of the active materials, interfaces and contacts. However, this can be neglected if the system is properly encapsulated. Diversely, degradation pathways due to light exposure and temperature cannot be eliminated and in general they might induce chemical reactions, interface degradations and morphological changes within the OPV stack [8]. In particular, the chemical and physical stability of the BHJ active layer is one of the most critical aspects, as one major problem associated with the application of polymers is their vulnerability under use conditions. The incident light is one of the main factors responsible for polymer degradation. Various reactions generate this instability involving rearrangements, chain scissions and crosslinking of the chemical structures, thus determining the loss of  $\pi$ -conjugation with a consequent bleaching, as

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evidenced by a typical decrease in intensity of the main absorption bands of the semiconducting materials [9].

To gain deeper insight into the intrinsic stability of  $\pi$ -conjugated polymers for OPV, several studies that compare the photostability of various materials in air, by monitoring their UV–visible photo-bleaching as a function of degradation time, have been published during the last decade [10]. Note that, the photochemically induced degradation processes that take place in air are significantly faster than those occurring when the irradiation is carried out on materials in the absence of oxygen [9,11].

The polymer stability and the rate of photodegradation are strongly dependent on the nature of the polymer structure, indeed slight structural changes can result in huge variations in the photochemical stability. Moreover, it was demonstrated that the number, size and nature of the side-chains, which are fundamental to tune several properties such as: i) solubility and processability, ii) position of the HOMO and LUMO levels, iii) structural conformation and iv) intra- and inter-chain interactions of the polymer chains, also play a key role in polymer photodegradation [12].

It is therefore crucial to further investigate the effect of the structural tuning of the side chains on the stability of the corresponding semiconducting polymers in order to generate knowledge and guidelines for the design of improved donor materials for efficient and photochemically stable BHJ solar cells. To this end, the effect of polymer side chain manipulation on the photochemical stability of the resulting films has been here investigated and then correlated with the photovoltaic performance and lifetime measurements of the corresponding OPV devices.

We present a series of p-type copolymers, P(1)–PFQBDT-OR, P(2)–PFQBDT-TROR, P(3)–PFQBDT-TR and P(4)–PFQTT (abbreviated in the text as P(1), P(2), P(3), and P(4), respectively), which combine a fluorinated quinoxaline (FQ) acceptor unit either with a differently substituted benzodithiophene (BDT) or an unsubstituted thieno[3,2-b]thiophene (TT) (Fig. 1). This work describes the synthesis and optical, electrochemical and photovoltaic properties of the polymers and investigates how different side chains (or absence as in the case of the reference TT-based polymer, P(4)) on the BDT unit influence the photostability of the resulting thin films. We firstly investigated the intrinsic stability of each polymer by monitoring the UV–vis absorption decay of the corresponding neat films, under 1 sun illumination, as a function of ageing time.

Next, BHJ solar cells using P(1)–P(4) as donor materials were fabricated and tested during prolonged light exposure in order to evaluate and compare the photostability responses of more complex systems such as complete polymer solar cells, under working conditions.

## 2. Experimental section

### 2.1. Electrochemical properties

Square-wave voltammetric measurements were carried out on a CH-Instruments 650A Electrochemical Workstation. A three-electrode setup consisting of platinum wires, both as working electrode and counterelectrode, and a Ag/Ag+ quasi-reference electrode were used. A 0.1 M solution of tetrabutylammonium-hexafluorophosphate ( $\text{Bu}_4\text{NPF}_6$ ) in anhydrous acetonitrile was used as supporting electrolyte. The polymers were deposited onto the working electrodes from chloroform solutions. The electrolyte was bubbled with nitrogen gas prior to each experiment. During the scans, nitrogen gas was flushed over the electrolyte surface. After each experiment, the system was calibrated by measuring the ferrocene/ferrocenium ( $\text{Fc}/\text{Fc}^+$ ) redox peak. The HOMO and LUMO energy levels of the polymers and electron acceptors were calculated from the peak values of the third scans by setting the oxidative peak potential of  $\text{Fc}/\text{Fc}^+$  vs the normal-hydrogen electrode (NHE) to 0.630 V and the NHE vs the vacuum level to 4.5 V [13].

### 2.2. Preparation and photodegradation of neat polymer films

Each polymer was dissolved in chloroform (with a concentration of 10 mg/ml) and then spin-coated on cleaned glass substrate (thickness of  $\sim 80$  nm for all films). Then, the samples were illuminated at 1 sun in air using an open array solar simulation system from Eye Lighting International (AM1.5G spectral matching filter,  $100 \text{ mW}/\text{cm}^2$ ). The samples were removed periodically to record the UV–vis absorbance spectra at room temperature with a Perkin Elmer Lambda 900 spectrophotometer. The thickness of the various films was measured by a profilometer (KLA Tencor, P-6).

### 2.3. OPV device fabrication and characterization

All materials, PEDOT:PSS (poly(3,4-ethylenedioxythiophene):poly(4-styrenesulfonate), Clevios P VP A1 4083, H.C. Starck),  $\text{PC}_{61}\text{BM}$  ([6,6]-phenyl-C61-butyric acid methyl ester, Solenne BV) and anhydrous *o*-dichlorobenzene (ODCB) were purchased from commercial sources (Sigma-Aldrich) and used without further purification. Patterned ITO-coated glasses ( $R_s \sim 10 \Omega/\text{sq}$ ) were cleaned in sequential sonicating baths (for 15 min) of deionized water, acetone and isopropanol. After the final sonication step, substrates were dried with a stream of  $\text{N}_2$  gas and then placed in an oxygen plasma chamber for 10 min. Next, a thin layer ( $\sim 30$  nm) of PEDOT:PSS was blade-coated on the ITO surface and subsequently annealed at  $150^\circ\text{C}$  for 15 min. The active layer blend solutions were formulated inside the glove box and stirred overnight at  $80^\circ\text{C}$ . For optimized devices, a total concentration of 30 mg/mL in ODCB was used for all donor polymers. A small percentage (3%, v/v) of solvent additive 1,8 diiodooctane (DIO) was added to the blend solutions of P(2) and P(4), to improve the thin film quality and the OPV performance of the corresponding devices. The blend solutions were deposited in air by blade-coating on top of the ITO/PEDOT:PSS surface. Before cathode deposition, the substrates were then either thermally annealed or left as-cast. To complete the device fabrication, LiF/Al (0.6 nm and 100 nm) were deposited sequentially without breaking vacuum ( $\sim 1 \times 10^{-6}$  Torr)

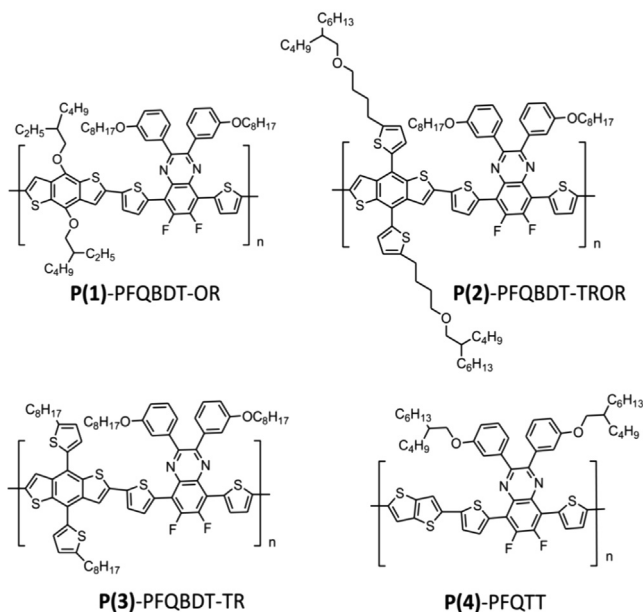


Fig. 1. Chemical structures of polymers P(1)–P(4).

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