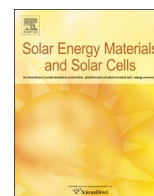




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## Enhanced thermal stability of organic photovoltaics via incorporating triphenylamine derivatives as additives

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## ABSTRACT

In this work, we prepared four star-shaped conjugated small molecules, the triphenylamine dithiophene (TBT) derivatives, namely TBT-H, TBT-Br, TBT-OH, and TBT-N<sub>3</sub> presenting hydride, bromide, hydroxyl, and azide terminal functional groups, respectively. These TBT derivatives were used as additives in the active layers of organic photovoltaics to investigate the effect of intermolecular interactions (TBT-H, TBT-OH) or crosslinking (TBT-N<sub>3</sub>, TBT-Br) on the long-term thermal stability of the devices. From analyses of blend film morphologies, and optoelectronic and device performance, we observed significant enhancements in thermal stability during accelerated heating tests at 150 °C for the devices incorporated with the additives TBT-N<sub>3</sub> and TBT-Br. These two additives functioned as crosslinkers, and constructed local borders that effectively impeded heat-promoted fullerene aggregation, thereby leading to highly stable morphologies. When compared with corresponding normal devices, the TBT-N<sub>3</sub>-derived devices based on poly(3-hexylthiophene) exhibited greater stability, with the power conversion efficiency (PCE) remaining as high as 2.5% after 144h at 150 °C. Because of this enhancement, a device based on an amorphous low-bandgap polymer, namely poly(thieno[3,4-*b*]thiophene-*alt*-benzodithiophene), with the addition of TBT-N<sub>3</sub> was fabricated. We observed a significant improvement in device stability, retaining approximately 60% (from 5.0 to 3.3%) of its initial PCE under accelerated heating (150 °C). In contrast, the PCE of the corresponding normal device decayed to 0.01% of its initial value.

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## 1. Introduction

Organic photovoltaics (OPVs) based on solution-processed bulk-heterojunction (BHJ) blends continue to attract attention because of their flexibility, light weight, low cost, short payback time, large-area fabrication, and amenability to roll-to-roll processing [1–5]. Many enhancements in device performance have been demonstrated through advances in material design and device engineering; these efforts have led to power conversion efficiencies (PCE) of up to 10%, the watershed for manufacturability [6–11]. Despite such high performance, for practical applications the devices must be stable when operated at elevated temperatures under constant thermal cycles and continuous illumination [12]. The stability of an OPV device can be correlated to its degree of encapsulation, the photo-degradation of its organic materials [13], the interfacial properties of each layer, and the morphology of

its blends. Each of these characteristics requires detailed investigation to determine its effect on the stability of OPV devices. In this study, we focused on the thermal stability and morphology of the blends under accelerated high temperature conditions.

Active layers composed of poly(3-hexylthiophene) and phenyl-C<sub>61</sub>-butyric acid methyl ester (P3HT:PC<sub>61</sub>BM) remain the standard research and industry system used to manufacture model OPVs, due to their reliable material sources, favorable performance-to-cost ratios, amenability to large-scale production, and ready processing in air [14,15]. In addition, a BHJ blend based on a bicontinuous interpenetrating network requires specific nano-sized phase segregation (on the order of the exciton diffusion length, 10–30 nm) to achieve the optimized morphology for efficient charge transfer, transportation in the blend film, and collection at the respective electrodes [16,17]. Thus, the major problem affecting the commercialization of OPVs is whether the highly efficient devices can be stabilized for long periods of time and, if so, whether the formulations empowering this morphological stability can be easily manipulated. The optimal morphology of an OPV is metastable; its blend must be capable of spontaneously moving to a low activation energy state to achieve

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thermodynamic stability. Conjugated polymers tend to undergo high degrees of molecular packing or crystallization, while fullerenes tend to aggregate or grow into micro-sized crystals. Hence, high-performance devices tend to display insufficient thermal stability, and therefore have yet to attain commercialization [18]. Durable OPV devices have been prepared by taking advantage of crosslinking agents or intermolecular interactions (typically hydrogen bonding) to fix the BHJ morphology; for example, polymer additives (crosslinked with themselves or with the acceptor PCBM) [19–23], fullerene additives (crosslinked with PCBM) [24–26], certain additives that form secondary bonds to the fullerene acceptors or to themselves [27–29], and small-molecule crosslinkers [30–32]. These approaches can significantly impact the morphological stability, confirming that linking molecules through chemical bonds can be an efficient means of preventing aggregation of either the conjugated polymer or PCBM. Because of their ease of synthesis and well-defined structures, small molecular additives have attracted much attention. Wantz et al. demonstrated OPVs that were thermally stable at 150 °C, based on an additive, 4,4'-bis(azidomethyl)-1-1'-biphenyl (BABP) bearing a bis-azide at both ends of the molecule [30]. More recently, McCulloch et al. used an azide functionalized additive, 1,6-diazidohexane (DAZH), which undergoes crosslinking under UV irradiation, to realize a polysilaindacenodithiophene-benzothiadiazole (SiIDT-BT)-based device exhibiting a PCE of 4.1% that was stable for over 130 h when heated at 85 °C [32]. In a previous study, we reported a crosslinkable open-cage fullerene additive that functioned efficiently to stabilize the BHJ morphology and OPV performance, which remained close to 95% of its initial PCE under accelerated thermal conditions at 150 °C [26]. In this study, we synthesized TBT derivatives presenting various terminal functional groups, namely hydride, bromide, hydroxyl, and azide groups, and examined the utility of these molecules as additives for OPV applications [Fig. 1(a)]. TBT-H functioned as the model compound. As displayed in Fig. 1(b), TBT-OH (with hydroxyl end groups) allowed hydrogen bonding interactions to exist within the active layers. TBT-Br and TBT-N<sub>3</sub> featured reactive terminal groups and, therefore, behaved as crosslinking-type additives: TBT-Br for UV-induced self-crosslinking and TBT-N<sub>3</sub> for crosslinking with PCBM at high temperature through cycloaddition and the formation of five-membered-ring heterocycles. The effects of the terminal

functional groups of these TBT derivatives were investigated. TBT-N<sub>3</sub>-derived P3HT devices exhibited PCEs as high as 2.5% with excellent thermal stability for 144 h under accelerated heating conditions at 150 °C. In addition, a poly(thieno[3,4-*b*]thiophene-*alt*-benzodithiophene) (PTB7)-derived device incorporated with TBT-N<sub>3</sub> as an additive exhibited significant enhancement in device stability, retaining 60% of its original PCE after 18 h of thermal annealing at 150 °C; in comparison, the PCE of the corresponding normal device without the additive dropped dramatically from  $5.26 \pm 0.05$  to  $0.01 \pm 0.00\%$ .

## 2. Experimental section

### 2.1. Measurements

Current–voltage (*I*–*V*) curves of the OPV devices were measured using a computer-controlled Keithley 2400 source measurement unit (SMU) and a Newport solar simulator (Oriel Sol2A Class ABA Solar Simulators) under AM 1.5 G illumination ( $100 \text{ mW cm}^{-2}$ ). The illumination intensity was calibrated using a standard Si photodiode detector equipped with a KG-5 filter. The output photocurrent was adjusted to match the photocurrent of the Si reference cell to obtain a power density of  $100 \text{ mW cm}^{-2}$ . After encapsulation, all devices were operated under an ambient atmosphere at 25 °C.

### 2.2. Cell fabrication and testing

All BHJ OPV cells were prepared using the following device fabrication procedure: The glass/indium tin oxide (ITO) substrates [Sanyo, Japan ( $8 \Omega/\square$ )] were sequentially patterned lithographically, cleaned with detergent, ultrasonicated in acetone and isopropyl alcohol, dried on a hot plate (120 °C, 5 min), and treated with oxygen plasma for 5 min [33]. Poly(3,4-ethylene-dioxythiophene):polystyrenesulfonate (PEDOT:PSS, Baytron P-VP AI4083) was passed through a 0.45- $\mu\text{m}$  filter before being deposited through spin-coating (3000 rpm) in air onto the ITO at a thickness of approximately 30 nm, then dried at 150 °C for 30 min inside a glove box. A blend of PCBM and P3HT [0.9:1 (w/w),  $18 \text{ mg mL}^{-1}$  in *o*-DCB] was stirred overnight in *o*-DCB, filtered through a 0.2- $\mu\text{m}$  polytetrafluoroethylene (PTFE) filter, and then spin-

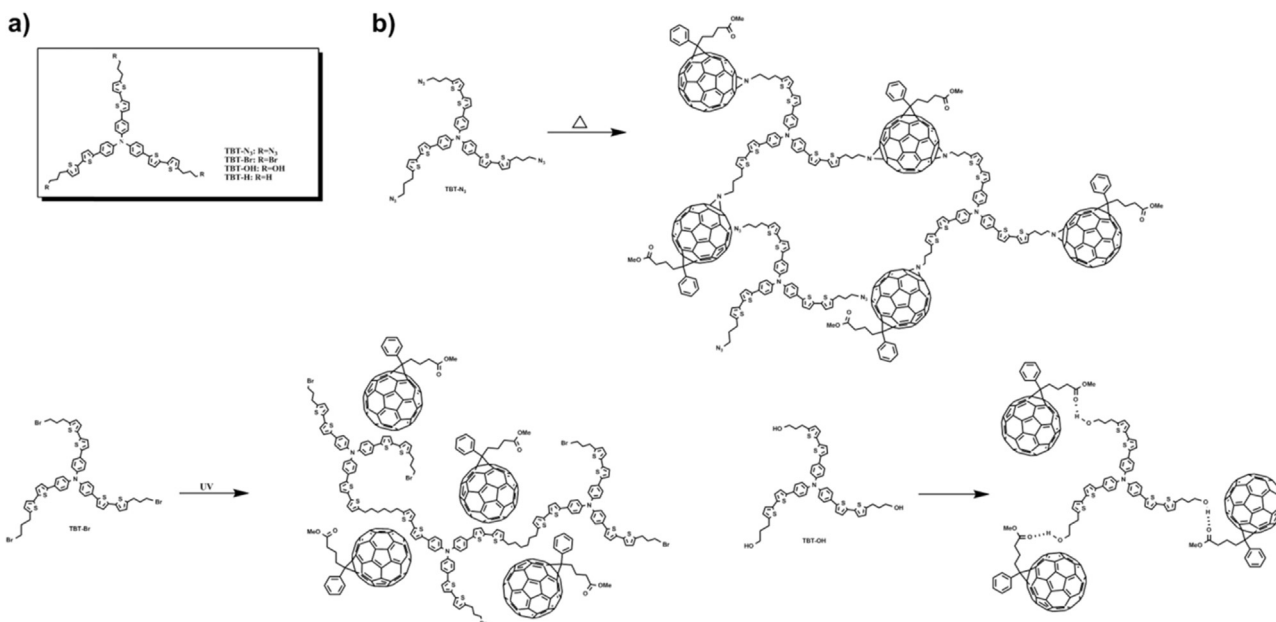


Fig. 1. (a) Chemical structures of TBT derivatives. (b) Possible interaction pathways for these additives within P3HT:PCBM blends.

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