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### **Paper:**

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# Advanced Materials

## An efficient, 'burn in' free organic solar cell employing a non-fullerene electron acceptor --Manuscript Draft--

<b>Manuscript Number:</b>	adma.201701156R1
<b>Full Title:</b>	An efficient, 'burn in' free organic solar cell employing a non-fullerene electron acceptor
<b>Article Type:</b>	Communication
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<b>Keywords:</b>	non-fullerene acceptor; Organic solar cells; Charge separation; burn in losses; trap assisted recombination
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<b>Additional Information:</b>	
<b>Question</b>	<b>Response</b>
<p>Please submit a plain text version of your cover letter here.</p> <p><b>If you are submitting a revision of your manuscript, please do not overwrite your original cover letter. There is an opportunity for you to provide your responses to the reviewers later; please do not add them here.</b></p>	<p>Dear Prof. Gregory,</p> <p>Please consider the manuscript entitled "An efficient, 'burn in' free organic solar cell employing a non-fullerene electron acceptor", co-authored by Hoyjung Cha, Jiaying Wu, Andrew Wadsworth, Jade Nagitta, Derya Baran, Iain McCulloch, James R. Durrant, for publication in Advanced Materials. The co-authors have all agreed to submission. The work has not been submitted for publication elsewhere.</p> <p>One of the biggest challenges limiting the commercial application of organic solar cells is that many, if not all, efficient organic solar cells to date exhibit a significant 'burn in' efficiency loss when first placed under solar irradiation, even when encapsulated against environmental ingress. Non-fullerene electron acceptors are attracting extensive interest as replacements for soluble fullerene derivatives such as PC61BM and PC71BM for organic electronics applications, including in particular organic bulk heterojunction solar cells. Studies of the application of NFA's to organic solar cells to date have largely focused on efficiency optimisation. We report herein that replacement of PCBM in an organic solar cell with a particular NFA, EH-IDTBR, results in devices, which are not only efficient but remarkably stable, exhibiting essentially no burn in loss of efficiency.</p> <p>In more detail, in this letter we focus on a comparison of the efficiency, stability and photophysics of organic solar cells employing PffBT4T-2OD as a donor polymer blended with either the non-fullerene acceptor EH-IDTBR or the fullerene derivative PC71BM as electron acceptors. Inverted PffBT4T-2OD:EH-IDTBR blend solar cells fabricated without any processing additive achieve power conversion efficiencies (PCEs) of <math>9.5 \pm 0.2</math> % and remarkably high open circuit voltages of <math>1.08 \pm 0.01</math> V. Photoluminescence quenching and transient absorption data are employed to elucidate the ultrafast kinetics and efficiencies of charge separation in both blends, with PffBT4T-2OD exciton diffusion kinetics within polymer domains, and geminate recombination losses following exciton separation being identified as key factors determining the efficiency of photocurrent generation. Remarkably, whilst encapsulated PffBT4T-2OD:PC71BM solar cells show significant efficiency loss under simulated solar irradiation ('burn in' degradation), PffBT4T-2OD:EH-IDTBR solar cells shows negligible burn in efficiency loss, as well as enhanced thermal stability. The greater susceptibility of the PCBM devices to burn in efficiency loss is correlated with a great susceptibility to the photo-generation of electronic trap states, reducing charge collection efficiency. This demonstration of an efficient, burn-in free organic solar cell is, we believe, a significant advance for the technological application of organic solar cells, and furthermore demonstrates a key advantage of non-fullerene acceptors in these devices. Consequently, we believe our work is appropriate for publication in Advanced Materials.</p> <p>We would like to propose the following as referees of this letter:</p> <p>Prof. Michael Chabinyk (UC Santa Barbara, USA), email:</p>

	<p>mchabinyc@engineering.ucsb.edu</p> <p>Prof. Kwang-Hee Lee (GIST, South Korea), email: klee@gist.ac.kr</p> <p>Prof. Feng Gao (Linkoping University, Sweden) email: fenga@ifm.liu.se</p> <p>Yours sincerely,</p> <p>James Durrant</p>
Do you or any of your co-authors have a conflict of interest to declare?	No. The authors declare no conflict of interest.
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<b>Order of Authors Secondary Information:</b>	
<b>Abstract:</b>	<p>We report a comparison of the efficiency, stability and photophysics of organic solar cells employing PffBT4T-2OD as a donor polymer blended with either the non-fullerene acceptor EH-IDTBR or the fullerene derivative PC71BM as electron acceptors. Inverted PffBT4T-2OD:EH-IDTBR blend solar cell fabricated without any processing additive achieve PCEs of <math>9.5 \pm 0.2</math> %. The devices exhibit a high open circuit voltage of <math>1.08 \pm 0.01</math> V, attributed to the high LUMO level of EH-IDTBR. Photoluminescence quenching and transient absorption data are employed to elucidate the ultrafast kinetics and efficiencies of charge separation in both blends, with PffBT4T-2OD exciton diffusion kinetics within polymer domains, and geminate recombination losses following exciton separation being identified as key factors determining the efficiency of photocurrent generation. Remarkably, whilst encapsulated PffBT4T-2OD:PC71BM solar cells show significant efficiency loss under simulated solar irradiation ('burn in' degradation) due to the trap-assisted recombination through increased photoinduced trap states, PffBT4T-2OD:EH-IDTBR solar cell shows negligible burn in efficiency loss. Furthermore, PffBT4T-2OD:EH-IDTBR solar cells were found to be substantially more stable under 85°C thermal stress than PffBT4T-2OD:PC71BM devices.</p>

DOI: 10.1002/adma.201701156

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2 **Communication**  
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6 **An efficient, 'burn in' free organic solar cell employing a non-fullerene electron acceptor**  
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9 *Li<sup>3</sup>, Justin Searl<sup>3</sup>, Mark F. Wyatt<sup>4</sup>, Derya Baran<sup>5</sup>, Ji-Seon Kim<sup>2</sup>, Iain McCulloch<sup>1,5</sup>, James R.*  
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42 **Keywords:** non-fullerene acceptor; organic solar cells; charge separation; burn in losses; trap assisted  
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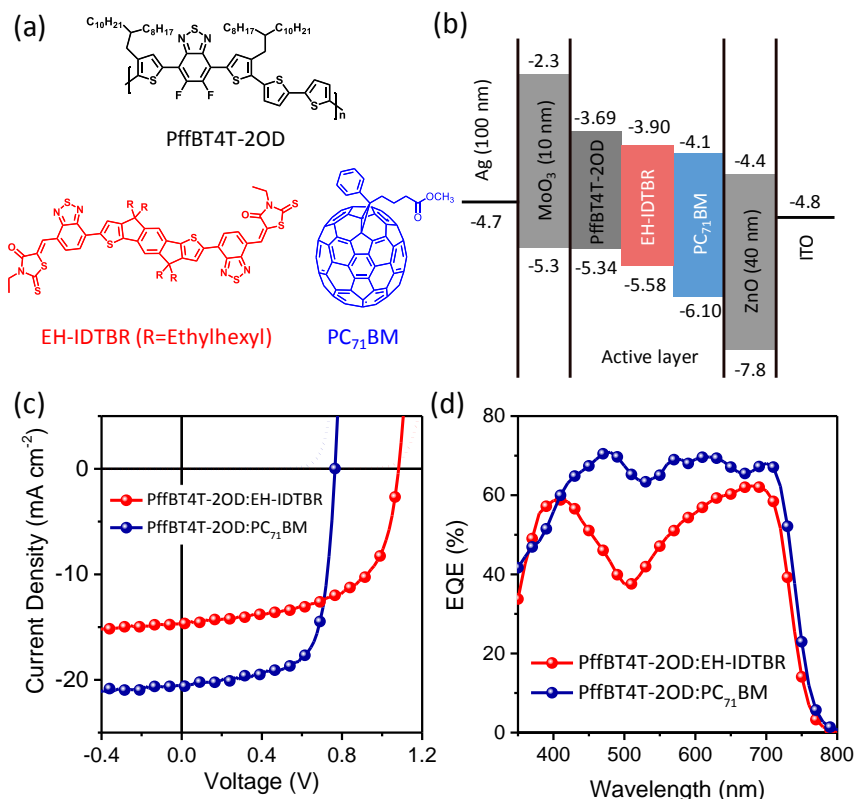
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48 Organic solar cells (OSCs) require both efficient and stable operation to become viable for commercial  
49 application.<sup>[1]</sup> However, many organic photoactive materials and blends are susceptible to a range of  
50 light induced degradation mechanisms which often results in a significant decrease in the solar cell  
51 performance under solar irradiation.<sup>[2]</sup> In particular, bulk heterojunction OSCs incorporating C<sub>60</sub>-based  
52 fullerene electron acceptors often show a significant initial loss of performance under sunlight  
53 exposure on a timescale of tens of hours before stabilising at an efficiency 25-50% lower than the  
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1 initial value – a loss of performance widely referred to as ‘burn in’.<sup>[3]</sup> Several possible origins of burn-  
2 in efficiency loss have been discussed in the literature,<sup>[2]</sup> including metal ion migration from device  
3 contacts,<sup>[4]</sup> photo-induced fullerene dimerization,<sup>[5]</sup> the photo-generation of electronic trap states<sup>[6]</sup> and  
4 most recently spinodal demixing.<sup>[7]</sup> Thermally induced performance degradation has also been  
5 identified as significant issue for many OSCs and attributed primarily to thermally induced fullerene  
6 aggregation/crystallization; under modest thermal stress conditions, limited photoinduced fullerene  
7 dimerization has been reported to be beneficial for device stability.<sup>[8]</sup> A further stability consideration is  
8 the use of process additives such as diiodooctane (DIO), such additives are widely used to increase the  
9 efficiency of polymer:fullerene solar cells, but have also been shown to impact detrimentally on device  
10 stability.<sup>[9]</sup> As such the development of efficient organic solar cells which exhibit stable performance  
11 without significant burn-in loss remains a significant challenge in this field.  
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25 The last two years have seen rapid progress in the development of non-fullerene acceptors (NFAs) for  
26 bulk heterojunction organic solar cells, yielding device efficiencies over 11%.<sup>[10]</sup> Such NFAs can  
27 exhibit several potentially attractive features for organic solar cell application: strong and tunable light  
28 absorption, tunable molecular energy levels and lower cost synthesis.<sup>[10]</sup> Given the greater synthetic  
29 flexibility of NFAs compared to fullerene acceptors, it is interesting to consider whether such OSCs  
30 employing NFAs can exhibit reduced burn-in degradation losses compared to fullerene acceptors, a  
31 subject to our knowledge not yet addressed in the literature to date.  
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42 We have recently reported a range of rhodanine-based NFAs which have achieved very promising  
43 device efficiencies in blends with P3HT, with efficiencies for ternary blends of up to 7.7%.<sup>[12-15]</sup> Such  
44 blends with P3HT have also shown promising unencapsulated device stabilities, significantly  
45 improved compared to those of analogous PC<sub>61</sub>BM devices, and indicative of promising resistance to  
46 oxygen and water induced degradation.<sup>[13]</sup> We have moreover shown that the replacement of P3HT  
47 with a lower bandgap highly crystalline polymer, PffBT4T-2DT can result in further improvements in  
48 device performance, with power conversion efficiencies approaching 10 % and high open circuit  
49 voltages including very low voltage loss < 0.5 V.<sup>[15]</sup> In this report, we focus on a comparison of blends  
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1 of an analogous crystalline donor PffBT4T-2OD<sup>[16]</sup> with PC<sub>71</sub>BM and with the non-fullerene small  
2 molecule EH-IDTBR. Inverted PffBT4T-2OD:EH-IDTBR blend solar cell fabricated without any  
3 processing additive achieve PCEs of  $9.5 \pm 0.2\%$ . The devices exhibit a high open circuit voltage of  
4  $1.08 \pm 0.01$  V, attributed to the high LUMO level of EH-IDTBR. Photoluminescence quenching and  
5 transient absorption data are employed to elucidate the ultrafast kinetics and efficiencies of charge  
6 separation in both blends, with PffBT4T-2OD exciton diffusion kinetics within polymer domains, and  
7 geminate recombination losses following exciton separation being identified as key factors  
8 determining the efficiency of photocurrent generation. Remarkably, whilst encapsulated PffBT4T-  
9 2OD:PC<sub>71</sub>BM solar cells show significant efficiency loss under simulated solar irradiation ('burn in'  
10 degradation) due to the trap-assisted recombination through increased photoinduced trap states,  
11 PffBT4T-2OD:EH-IDTBR solar cell shows negligible burn in efficiency loss. Furthermore, PffBT4T-  
12 2OD:EH-IDTBR solar cells are found to be substantially more stable under 85°C thermal stress than  
13 PffBT4T-2OD:PC<sub>71</sub>BM devices.  
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**Figure 1.** (a) Chemical structures of PffBT4T-2OD as an electron acceptor and EH-IDTBR and PC<sub>71</sub>BM as electron acceptors, (b) energy levels of the materials used in this work from thin films [13,14]; HOMO levels measured by cyclic voltammetry and LUMO levels calculated based on HOMO levels and optical bandgaps, (c) the  $J$ - $V$  curve of PffBT4T-2OD-based solar cells, and (d) the EQE curve of PffBT4T-2OD-based solar cells.

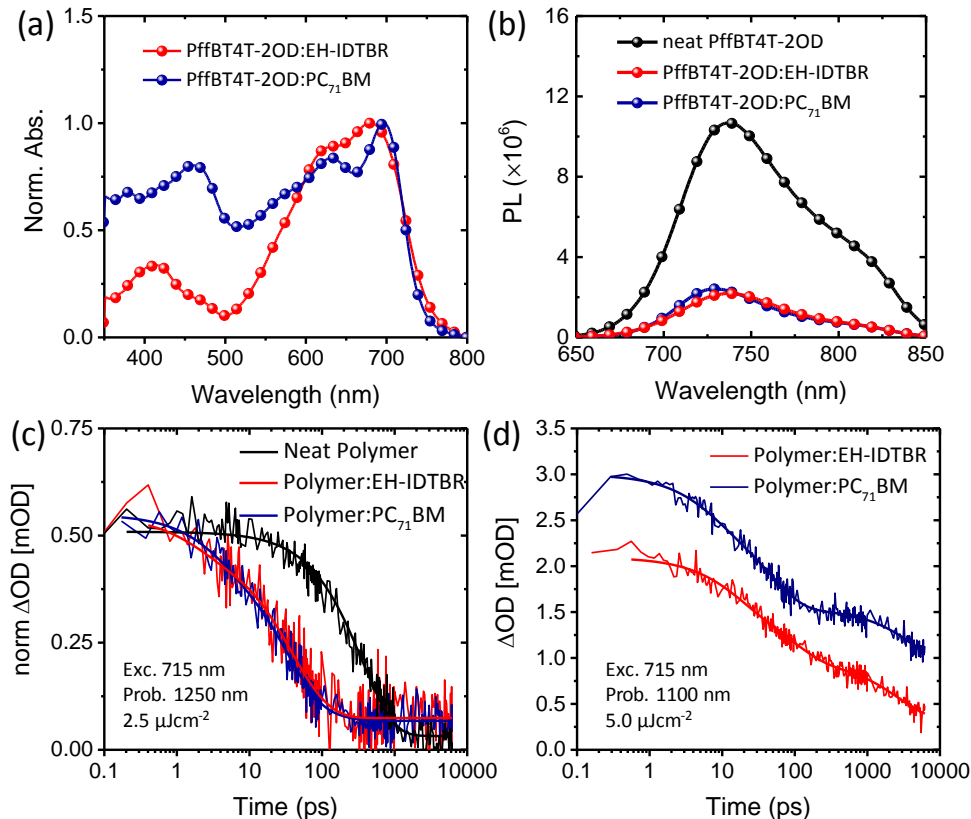
The bulk heterojunction solar cells employed in this study use a non-fullerene small molecule EH-IDTBR<sup>[12-14]</sup> as the electron acceptor; for the donor, we employed a commercially available low bandgap polymer, PffBT4T-2OD,<sup>[16]</sup> which has high crystallinity and high molecular weight ( $M_n > 54,900$ , PDI = 2.14) (**Figure 1a**). We also prepared PffBT4T-2OD:PC<sub>71</sub>BM-based solar cells as control samples. The devices were fabricated in an inverted configuration, using ZnO as an electron transport layer and MoO<sub>3</sub> as a hole transport layer (Figure 1b). For an optimised combination of PffBT4T-2OD and EH-IDTBR blend in the active layer, we adjusted the thermal annealing temperature and modified the film morphology with the various solvents (Table S1). For devices based on EH-IDTBR, we obtained optimal device performance with donor:acceptor mass ratio of 1:1.4 in dichlorobenzene without any processing additives (tests with processing additives did not

1 result in any significant improvement in device performance). In contrast, efficient device  
2 performance was only obtained for PffBT4T-2OD:PC<sub>71</sub>BM devices using a  
3 chlorobenzene:dichlorobenzene (1:1) mixed solvent in the presence of the processing additive  
4 diiodooctane (DIO: a high boiling point solvent in which PC<sub>71</sub>BM is highly soluble), as reported  
5 previously.<sup>[16]</sup> The current density voltage (*J-V*) curves for optimised PffBT4T-2OD:EH-IDTBR and  
6 PffBT4T-2OD:PC<sub>71</sub>BM solar cells are shown in Figure 1c. The device parameters are listed in Table  
7 S1, including consideration of performance reproducibility. Overall the PffBT4T-2OD:EH-IDTBR  
8 solar cell exhibit efficiencies up to  $9.5 \pm 0.2\%$ , whilst the PffBT4T-2OD:PC<sub>71</sub>BM solar cells exhibited  
9 efficiencies up to  $10.9 \pm 0.1\%$  with DIO. PffBT4T-2OD:PC<sub>71</sub>BM solar cells processed without DIO  
10 exhibited efficiencies of only up to 6.6% and poorer reproducibility, and will not be considered further  
11 herein. The devices employing EH-IDTBR exhibited higher voltages than device employing PC<sub>71</sub>BM,  
12 attributed primarily to the higher LUMO energy of EH-IDTBR, as we have discussed in more detail  
13 for analogous devices elsewhere,<sup>[15]</sup> but lower photocurrents, as discussed below.  
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30 The UV-Visible absorption spectra of the PffBT4T-2OD:EH-IDTBR and PffBT4T-2OD:PC<sub>71</sub>BM  
31 blends are shown in **Figure 2a**, respectively. Figure 1d displays the external quantum efficiency  
32 (EQE) spectra for both devices. PffBT4T-2OD:PC<sub>71</sub>BM based devices show broad and strong photo  
33 response from 350 to 800 nm, which is consistent with their broad UV-Vis absorption spectrum.  
34 Compared with the PffBT4T-2OD:PC<sub>71</sub>BM-based solar cells, the PffBT4T-2OD:EH-IDTBR solar  
35 cells show a significant decrease in the photo response at around 500 nm due to the lack of absorption  
36 of both photoactive materials. We also note that optimised EH-IDTBR devices have thinner film  
37 thicknesses than PC<sub>71</sub>BM (~70 and ~200 nm, respectively), which further contributes to the weak light  
38 absorption of the PffBT4T-2OD:EH-IDTBR solar cells around 500 nm. The integrated short circuit  
39 current ( $J_{SC}$ ) values from these EQE data for PffBT4T-2OD:PC<sub>71</sub>BM and PffBT4T-2OD:EH-IDTBR  
40 are 18.7 and 14.0 mAcm<sup>-2</sup>, respectively, in agreement (within 5% of the measured  $J_{SC}$  values. Overall,  
41 we can conclude that the more limited absorption range covered by the PffBT4T-2OD:EH-IDTBR  
42 blend, due to the overlap of the EH-IDTBR and PffBT4T-2OD absorption bands, is the primary cause  
43 of the lower  $J_{SC}$ . Despite this poorer light harvesting, it is striking that these devices still yield device  
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efficiencies of 9.5%, suggesting that further tuning of energy levels to yield complimentary absorption bands is a promising route to further enhance device efficiencies.



**Figure 2.** (a) Normalized UV-Visible absorption spectra and (b) PL spectra excited at 715 nm for neat PffBT4T-2OD and PffBT4T-2OD:PC<sub>71</sub>BM and PffBT4T-2OD:EH-IDTBR blend films. (c) Transient absorption data showing the time evolution of PffBT4T-2OD singlet exciton absorption at probed at 1250 nm for the corresponding films (normalized at 1 ps) and (d) showing the time evolution of singlet exciton (up to  $\sim 100$  ps) and polaron absorption (after  $\sim 100$  ps) for PffBT4T-2OD:PC<sub>71</sub>BM and PffBT4T-2OD:EH-IDTBR blends probed at 1100 nm (normalized for photons absorbed). The solid lines are the exponential decay fitting of absorption.

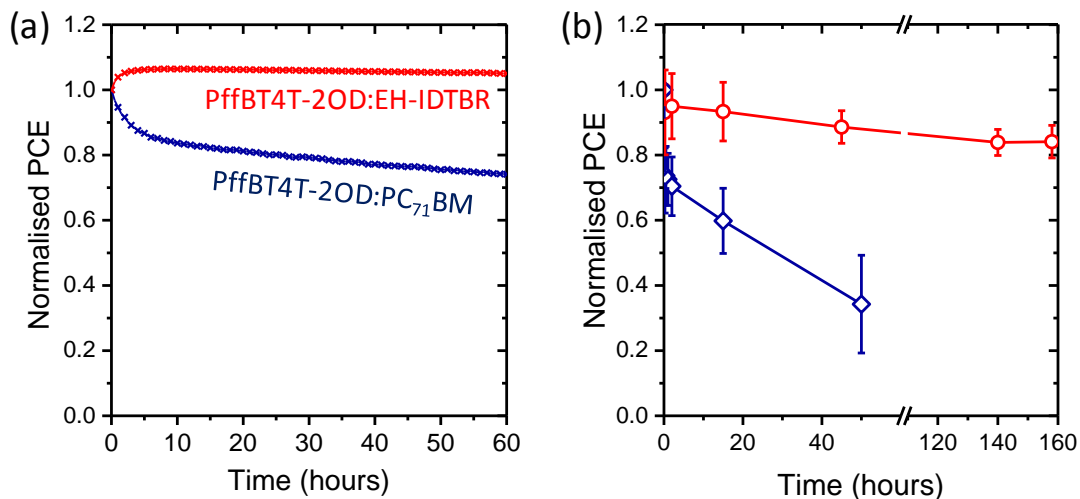
Photoluminescence (PL) studies were carried out on the PffBT4T-2OD:EH-IDTBR and PffBT4T-2OD:PC<sub>71</sub>BM blends as shown in Figure 2b. The polymer PL quenching efficiencies (PLQE) are similar for both blends: 80% for PffBT4T-2OD:EH-IDTBR and 77% for PffBT4T-2OD:PC<sub>71</sub>BM. These relatively modest PL quenching efficiencies can be attributed most probably to the high

1 crystallinity and large domain size of PffBT4T-2OD,<sup>[16]</sup> resulting in significant polymer exciton decay  
2 to ground during exciton diffusion to the polymer/acceptor interface. This exciton decay to ground  
3 corresponds to a ~20% quantum efficiency loss, and is likely to be the primary cause of the sub-unity  
4 maximal EQE data shown in Figure 1d.  
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10 To investigate further charge generation and recombination dynamics in the two blend films studied  
11 herein, femtosecond transient absorption spectroscopy (fs-TAS) was employed (Figures 2c and 2d).  
12 We used an excitation wavelength of 715 nm for both thin films, corresponding to the PffBT4T-2OD  
13 absorption maximum, although we note that EH-IDTBR also absorbs at this wavelength. Transient  
14 absorption spectra as a function of time delay for both blend films are shown in Figure S2, the  
15 corresponding control data for neat films are shown in Figure S1. Figure 2c plots the transient  
16 absorption decay dynamics of neat PffBT4T-2OD, PffBT4T-2OD:EH-IDTBR and PffBT4T-  
17 2OD:PC<sub>71</sub>BM films monitored at 1250 nm, assigned to the decay kinetics of PffBT4T-2OD singlet  
18 excitons (at this wavelength both EH-IDTBR excitons and the blend polarons yield relatively small  
19 signals, see SI for details). In the absence of acceptor, these excitons decay with a half-time of  $280 \pm$   
20  $10$  ps ( $2.5 \mu\text{J cm}^{-2}$ ), accelerating to half-times of  $30 \pm 0.5$  and  $28 \pm 0.5$  ps in blends with PC<sub>71</sub>BM and  
21 EH-IDTBR respectively. These accelerated exciton decays, assigned to exciton quenching due to  
22 electron transfer to the molecular acceptors, are in reasonable agreement with the PL quenching  
23 efficiencies measured above (PLQE of 77 and 80% for blends with PC<sub>71</sub>BM and EH-IDTBR,  
24 respectively). This agreement, and the observation of a single exciton decay phase, indicate PffBT4T-  
25 2OD in the blend films is primarily in pure polymer domains, with relatively little molecular scale  
26 mixing with the molecular acceptors. These data support our conclusion above that PffBT4T-2OD  
27 exciton decay to ground during diffusion within pure polymer domains results in a 10 - 20 % quantum  
28 efficiency loss in these devices.  
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53 Whilst the PffBT4T-2OD exciton separation kinetics are very similar for both blend films, the yields  
54 and lifetimes of the photogenerated charges resulting from this exciton separation are clearly distinct.  
55 This can observe from the amplitude of the PffBT4T-2OD polaron absorption observed centred at  
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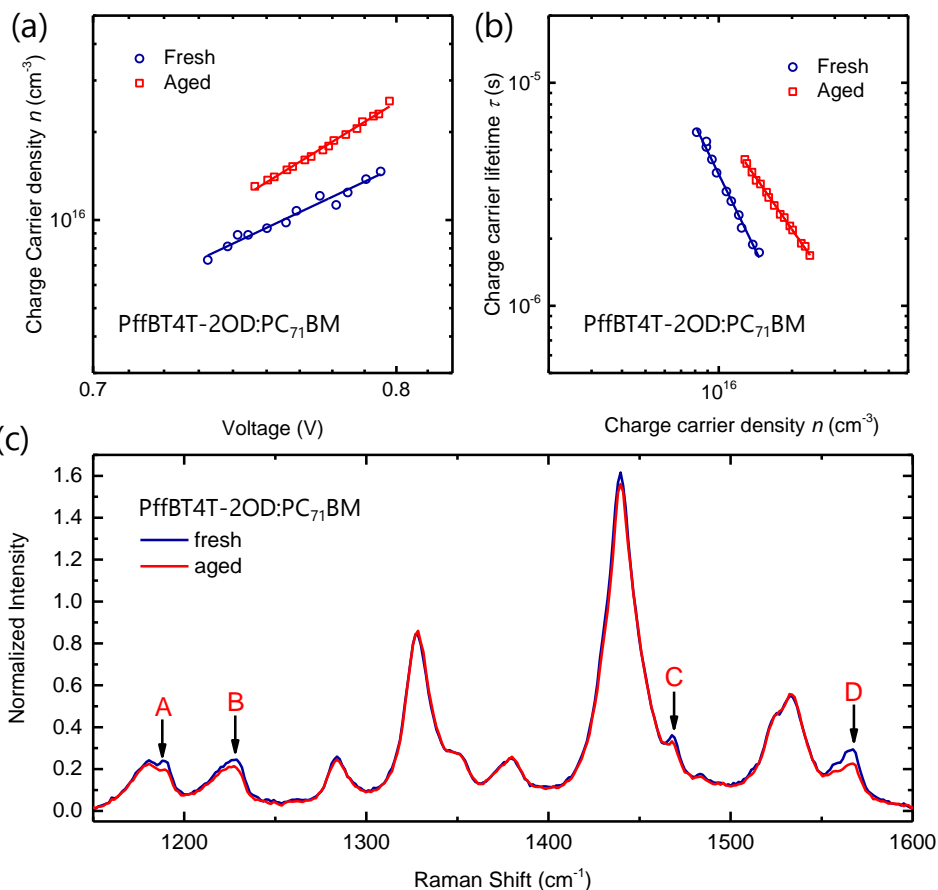
1 1100 nm at long time delays in both films in Figures S2a and S2b, and illustrated most clearly as the  
2 slow decay phase in the transient kinetics plotted at this wavelength in Figure 2d (the initial decay  
3 phase is assigned to the decay of exciton absorption). It is apparent that compared to PffBT4T-  
4 2OD:PC<sub>71</sub>BM blends, PffBT4T-2OD:EH-IDTBR blend shows a lower amplitude and shorter life-time  
5 for this polaron signal (this plot is normalized for matched densities of absorbed photons). For both  
6 blends, these polaron decays apparent from 1–6 ns were found to be excitation intensity independent  
7 (Figure S2) and therefore are assigned primarily to geminate recombination (at higher excitation  
8 densities, the decays become excitation density dependent, assigned to non-geminate recombination  
9 becoming faster than geminate recombination). It thus appears that geminate recombination losses are  
10 more significant for PffBT4T-2OD:EH-IDTBR than for PffBT4T-2OD:PC<sub>71</sub>BM. The importance of  
11 geminate (monomolecular) recombination losses rather than non-geminate (bimolecular)  
12 recombination losses in limiting  $J_{SC}$  was supported by plots of  $J_{SC}$  vs light intensity ( $I$ ), which indicate  
13 almost linear behavior for both devices ( $J_{SC} \sim I^\beta$ ,  $\beta \geq 0.98$  up to 1 sun). These increased geminate  
14 recombination losses most likely result from the high LUMO level of EH-IDTBR resulting in a lower  
15 energy offset driving charge separation (several studies have reported that lower energy offsets  
16 primarily cause on increased geminate recombination losses rather than slower or less efficient exciton  
17 separation).<sup>[17]</sup> Increased geminate recombination losses for the EH-IDTBR blend are likely to be an  
18 additional factor limiting photocurrent generation and EQE values for this devices fabricated with this  
19 blend, although a quantitative analysis of this issue would require consideration of the field  
20 dependence of these geminate recombination losses, beyond the scope of this study.  
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**Figure 3.** Normalised PCEs of the devices after long-term (a) light soaking without UV light maintained below 50°C thermal stress and (b) annealing at 85°C, under nitrogen.

We turn now to consideration of the stability of PffBT4T-2OD:PC<sub>71</sub>BM and PffBT4T-2OD:EH-IDTBR solar cells. Device stability was tested for glass encapsulated devices under light or thermal stress and unencapsulated devices under dark in air. Normalised power conversion efficiency (PCE) data are shown in **Figure 3**, with full device characteristics detailed in Figures S3 and S4. Under white light stress, the PCE of a representative PffBT4T-2OD:PC<sub>71</sub>BM-based device, decreased up over 20% over 60 hours, typical of light induced burn in losses reported for other OSC employing fullerene acceptors.<sup>[3]</sup> In contrast the PffBT4T-2OD:EH-IDTBR device shows essentially no light induced efficiency loss over this timescale, exhibiting a PCE of  $9.5 \pm 0.2\%$  after 60 hours under illumination indeed in some cases a modest increase in performance was observed, attributed to photoinduced doping of the ZnO layer.<sup>[18]</sup> This difference in burn-in response was tested for 18 devices, in all tests the PffBT4T-2OD:EH-IDTBR showed substantially less or negligible burn in efficiency loss compared to PffBT4T-2OD:PC<sub>71</sub>BM devices (normalized PCE losses after 60 hours of  $0 \pm 4\%$  and  $-26 \pm 8\%$  respectively). Under 85 °C thermal stress in the dark, the PffBT4T-2OD:EH-IDTBR device also showed substantially improved stability compared to PffBT4T-2OD:PC<sub>71</sub>BM devices (Figure 3b), with the EH-IDTBR showing a normalized PCE loss of only  $16 \pm 4\%$  after 160 hours of thermal loss,

1 while that of the PffBT4T-2OD:PC<sub>71</sub>BM based device dropped  $73 \pm 15\%$  within 60 hours. We note  
2 that the loss of device performance with PC<sub>71</sub>BM showed clearly different origins under light and  
3 thermal stress. Under light stress, PCE loss derived primarily from loss of fill factor (FF), indicative of  
4 increased charge trapping and recombination.<sup>[6]</sup> Under thermal stress, PCE loss derived primarily from  
5 a loss of  $J_{SC}$ , indicative of thermally induced phase segregation.<sup>[6,8]</sup> Supporting this conclusion for the  
6 origin of thermally induced efficiency loss, AFM data indicate a significant increase in film roughness  
7 following thermal stress for PffBT4T-2OD:PC<sub>71</sub>BM but not for PffBT4T-2OD:EH-IDTBR (see Figure  
8 S5), and optical microscopy images indicate the formation of micrometre-sized clusters after thermal  
9 stress at 85 °C for 12 hours for PffBT4T-2OD:PC<sub>71</sub>BM blend films, which were not observed for  
10 PffBT4T-2OD:EH-IDTBR blend films (see Figure S6). Consistent with this conclusion, we note that  
11 Brabec et al have recently reported that PffBT4T-2OD:PC<sub>61</sub>BM blends can be susceptible to  $J_{SC}$  loss  
12 due to spinodal demixing, consistent with the thermal degradation we observe herein but distinct from  
13 the FF loss we observe under light induced degradation.<sup>[7]</sup> We also tested the ambient shelf life  
14 stability of unencapsulated PffBT4T-2OD:EH-IDTBR and PffBT4T-2OD:PC<sub>71</sub>BM devices. Devices  
15 were stored at room temperature under dark conditions. After 95 h of air exposure PffBT4T-2OD:EH-  
16 IDTBR devices lost ~ 28% of their initial PCE, whereas PffBT4T-2OD:PC<sub>71</sub>BM devices performance  
17 lost 85% of their initial PCE after 65 h (Figure S7). These results suggest that PffBT4T-2OD:EH-  
18 IDTBR devices exhibit not only improved resistance to thermal and light stress, but also superior  
19 shelf-life performance under ambient exposure.



**Figure 4.** Transient optoelectronic analyses of PffBT4T-2OD:PC<sub>71</sub>BM device before and after light induced burn in efficiency loss. (a) Charge extraction (CE) measurements of charge carrier density at open circuit as a function of light intensity, plotted versus open circuit voltage; (b) the corresponding charge carrier lifetimes measured by transient photovoltage (TPV) decays, plotted against the measured charge densities. Device aging (burn in) was induced by the 540 nm light source employed in the CE and TPV measurements. (c) Representative Raman spectra of fresh and light-aged PffBT4T-2OD:PC<sub>71</sub>BM blend layer of devices, obtained via 488 nm laser excitation and normalized to polymer-only peak at 850  $\text{cm}^{-1}$ . Peaks labelled A-D are signatures of PC<sub>71</sub>BM. Light aging employed 4 hours LED white light irradiation as for Figure 3a.

Figure 3 indicates that PffBT4T-2OD:EH-IDTBR devices are significantly more stable under both light and thermal stress conditions than PffBT4T-2OD:PC<sub>71</sub>BM devices, and in particular show negligible burn in efficiency loss. The remarkable stability of PffBT4T-2OD:EH-IDTBR devices was

1 found to be rather specific to this combination of donor and acceptor materials, with for example,  
2 PffBT4T-2OD devices fabricated with some alternative rhodanine acceptors to EH-IDTBR showing  
3 greater burn-in efficiency losses, as we will report in detail elsewhere. We note that unlike the  
4 PffBT4T-2OD:PC<sub>71</sub>BM devices studied herein, the EH-IDTBR devices were fabricated without the  
5 DIO processing additive, which may in part explain the improved thermal stability. We also note that  
6 EH-IDTBR, in contrast to PC<sub>71</sub>BM, shows significant crystallinity,<sup>[13]</sup> which is also likely to be a  
7 factor in the improved thermal stability of EH-IDTBR devices.<sup>[1a]</sup>

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17 To investigate further the origin of the burn-in degradation observed herein, charge extraction (CE)  
18 and transient photovoltage (TPV) measurements were undertaken to determine the charge carrier  
19 densities and recombination dynamics of fresh and aged solar cells. For PffBT4T-2OD:PC<sub>71</sub>BM, these  
20 CE data, **Figure 4a**, indicate an ~ 50% increase in charge carrier density for matched cell open circuit  
21 voltage ( $V_{oc}$ )'s and therefore matched quasi-Fermi level splitting. As this charge carrier density is  
22 primarily in shallow trap states, this increase in charge density indicates that light induced burn in  
23 efficiency loss correlates with a 50% increase in the density of electronic trap states. This conclusion is  
24 supported by our observation of increased carrier lifetimes following burn in Figure 4b, indicative of  
25 increased charge trapping, and is also consistent with our observation that burn in efficiency loss  
26 primarily results from a loss of FF, indicative of a loss of collection efficiency. In contrast, for  
27 PffBT4T-2OD:EH-IDTBR devices (Figure S9), both CE and TPV measurements were observed to be  
28 relatively insensitive to device light exposure. These results were supported by ideality factor  
29 measurements, which indicate a modest increase in ideality factor for light aged PffBT4T-  
30 2OD:PC<sub>71</sub>BM devices, as expected for increased charge trapping following light exposure (Figure  
31 S8).<sup>[2d,6b]</sup>

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51 We turn now to the molecular origin of improved resistance of PffBT4T-2OD:EH-IDTBR devices  
52 to light induced charge trapping, and associated burn in performance loss. We note that, unlike  
53 PC<sub>61</sub>BM, PC<sub>71</sub>BM is resistant to photo-induced dimerization, so that differences in photo-induced  
54 dimerization are unlikely to explain the improved stability. AFM data indicates that the increase in  
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1 trap density for light exposed PffBT4T-2OD:PC<sub>71</sub>BM is not associated with a change in film  
2 morphology (Figure S10). In contrast, Raman spectra (Figure 4c) indicate that light exposure results in  
3 specific loss of some PC<sub>71</sub>BM Raman features, whilst those of PffBT4T-2OD are unchanged (see  
4 Raman spectra of neat PffBT4T-2OD and PC<sub>71</sub>BM films in Figure S11, respectively). In particular, 4  
5 hours LED light exposure results in 23% loss of a Raman feature at 1650 cm<sup>-1</sup> (marked as Feature D in  
6 Figure 4c) assigned previously to localized vibrations of C<sub>5</sub>/C<sub>6</sub> rings of the fullerene cage.<sup>[19]</sup> We note  
7 no frequency shifts in Raman features were observed, confirming negligible fullerene cage  
8 dimerization; similarly these Raman spectra rule out significant photooxidation of the C<sub>70</sub> cage.<sup>[20]</sup>  
9 Specific reduction of the 1650 cm<sup>-1</sup> Raman feature, with the other features being relatively invariant, is  
10 most likely due to light induced bond disruption or cleavage at the site of the solubilizing side group  
11 attachment to the C<sub>70</sub> cage. Whilst further work is clearly required to understand fully these  
12 observations, these data suggest that light induced burn in efficiency loss in the PffBT4T-  
13 2OD:PC<sub>71</sub>BM devices studied herein is associated with photoinduced degradation of PC<sub>71</sub>BM, with  
14 EH-IDTBR being less susceptible to such degradation.

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32 In summary, we conclude that the improved stability of PffBT4T-2OD:EH-IDTBR devices to light  
33 induced burn in efficiency loss is correlated with a greater resistance to photoinduced electronic trap  
34 state formation relative to devices employing PC<sub>71</sub>BM. We note that McGehee et al. have also  
35 observed that burn in efficiency losses are correlated with trap state formation and that these losses  
36 were reduced for more crystalline materials.<sup>[6]</sup> Clearly the stability data we report herein is limited in  
37 scope and timescale, and further work is required to elucidate fully the relationships between trap state  
38 formation, material structure and crystallinity, the use of processing additives and stability for the  
39 materials studied herein. However, the data herein demonstrate that, at least under the conditions  
40 studied, PffBT4T-2OD:EH-IDTBR devices show remarkably stability, significantly superior to that of  
41 PffBT4T-2OD:PC<sub>71</sub>BM devices, which is promising for future applications of this new NFA.

## 52 Supporting Information



Experimental detail, device optimisation, transient absorption spectra, photovoltaic results under the illumination, under thermal stress and in the dark under ambient conditions, AFM images, OM images transient optoelectronic analyses and Raman spectrum are included in Supporting Information. Supporting Information is available from the Wiley Online Library or from the author.

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**A high efficiency, burn in free non-fullerene-based PffBT4T-2OD:EH-IDTBR solar cell** is reported, fabricated without processing additives. Transient absorption and optoelectronic analyses elucidate the causes of this high efficiency and stability, with the superior stability compared to PC<sub>71</sub>BM devices being correlated with increased crystallinity and reduced photo-generation of trap states.

### Keyword

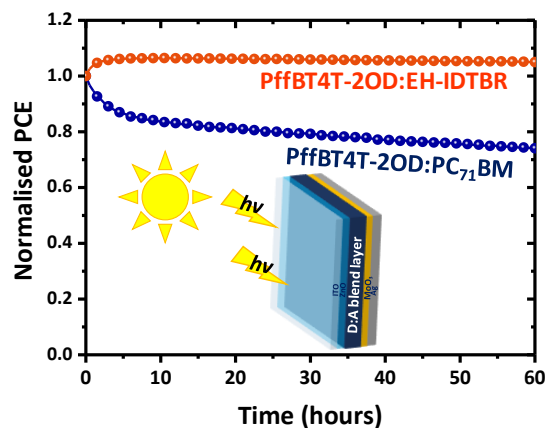
non-fullerene acceptor; organic solar cells; charge separation; burn in losses; trap assisted recombination

### Corresponding Author\*

Prof. J. R. Durrant

**An efficient, ‘burn in’ free organic solar cell employing a non-fullerene electron acceptor**

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