

# **2009 GCEP Report**

## **Project title: Advanced Materials and Devices for Low Cost and High Performance Organic Photovoltaic Cells**

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### **Abstract**

Understanding the loss mechanisms in current state-of-the-art organic photovoltaic devices is crucial to increasing their efficiency. Most high efficiency polymer:fullerene solar cells have external quantum efficiencies of only ~65%. By using light intensity to tune the steady-state concentration of carriers in the device, we show that the shape of the photocurrent versus voltage is independent of carrier concentration, which demonstrates that in these high efficiency cells there is no significant loss of carriers to bimolecular recombination or to space charge effects. This finding leads to the important result that matched charge carrier mobilities of  $\sim 3 \times 10^{-3} \text{ cm}^2/\text{V}\cdot\text{s}$  are sufficient to avoid these losses and should be used as design targets for new materials.

Exciton harvesting is of fundamental importance for the efficient operation of organic photovoltaic devices. The quantum efficiencies of many organic and hybrid organic-inorganic devices are still limited by low exciton harvesting efficiencies. This problem is most apparent in planar heterostructures that suffer from a direct tradeoff between light absorption and exciton harvesting. One approach to overcome small diffusion lengths is the use of triplet excitons. We investigated and analyzed solar cells from several materials. We conclude that triplet excitons do not provide any significant advantage over singlet excitons for several materials. While dilute guest:host configurations work well in OLEDs, where radiative recombination is desired and charge trapping is beneficial, solar cells require concentrated films. Because the triplet lifetime is shortened in such films, the triplet state is no longer advantageous.

We have found a general way to reduce bandgap of polymers while maintaining good charge transport properties. We have successfully synthesized and characterized a number of new low-band gap polymers. We achieved bandgap as low as 1.1 eV. Some of the polymers showed charge carrier mobility as high as  $0.2 \text{ cm}^2/\text{Vs}$  and power conversion efficiency of 1.6% illuminated by AM 1.5 light.

## Introduction

A significant fraction of the carbon released into the atmosphere is a result of burning coal and natural gas to produce electricity. It is therefore highly desirable to find ways to generate electricity without releasing carbon. The development of affordable photovoltaic (solar) cells is one of the most promising long-term solutions to keeping the CO<sub>2</sub> concentration in the atmosphere at safe levels.

Currently, almost all photovoltaic (PV) cells being manufactured are made of crystalline silicon. The average cost of the electricity that is generated in a sunny location using these cells is about two-four times more than the typical cost of electricity from the grid. Our goal is to develop technology that can reduce the cost per Watt of generation capability by at least a factor of five and hopefully even more. Our approach is to use organic semiconductors because they can be deposited onto flexible substrates in roll-to-roll coating machines, similar to those used to make photographic film and newspapers. In this project, we plan to work on three directions: (i) having significant exciton harvesting and preventing geminate recombination by using phosphorescent semiconductors, (ii) new device designs and materials required for double or triple the efficiency, and (iii) new materials design concepts to improve the absorption of low energy photons by using new low band gap semiconductors that have sufficient charge carrier mobilities for charge extraction.

## Results

### I. Analysis of Loss Mechanisms in Bulk Heterojunction Solar Cells

Understanding the loss mechanisms in current state-of-the-art organic photovoltaic devices is crucial to increasing their efficiency. Most high efficiency polymer:fullerene solar cells have external quantum efficiencies of only ~65%. By using light intensity to tune the steady-state concentration of carriers in the device, we show that the shape of the photocurrent versus voltage is independent of carrier concentration, which demonstrates that in these high efficiency cells there is no significant loss of carriers to bimolecular recombination or to space charge effects. This finding leads to the important result that matched charge carrier mobilities of  $\sim 3 \times 10^{-3}$  cm<sup>2</sup>/V.s are sufficient to avoid these losses and should be used as design targets for new materials. By comparing the shapes of the photocurrent curves for different device thicknesses, we show that the photocurrent scales with electric field. These findings strongly imply that losses are due to recombination of the geminate pairs, which leads to only ~75% and ~65% separation of these pairs under short circuit and maximum power conditions respectively. Applying a large reverse bias completely extracts all the geminate pairs, which strongly suggests that there are an insignificant number of dead-ends and disconnected islands in either phase in the polymer:fullerene nanostructure. By determining that geminate recombination is the most significant limiting factor in current high efficiency organic solar cells, this points us and other researchers to an issue that, once solved, should increase efficiency by a factor of ~30%

In this investigation, we systematically characterized all of the important properties of poly(3-hexyl thiophene) (P3HT) : [6,6]-phenyl C61-butyric acid methyl ester (PCBM)

bulk heterojunction solar cells. By evaluating all processes from light absorption to charge collection, we have painted a detailed picture of the important processes that together determine the overall efficiency. By modeling the optical interference effects inside the device, we determined that most of the light is absorbed by the polymer and that the overall efficiency is limited by an electronic process (characterized by the internal quantum efficiency). We performed photoluminescence quenching studies to show that exciton diffusion and harvesting and charge transfer are all close to 100% efficient. This finding is important because it implies that all of the inefficiencies are in charge collection, the process of the separated charges moving from the donor/acceptor interface to the electrodes. This process can be hindered by three types of recombination, geminate recombination, recombination due to space charge attenuation of the built-in field, and bimolecular recombination. Geminate recombination is recombination of the initially formed electron and hole across the donor/acceptor interface soon after the exciton is split. Bimolecular recombination is recombination of independent electrons and holes as they move through the bulk toward the electrodes. This type of recombination is dependent on the charge carrier concentration. Space charge build up is not an issue in bulk heterojunction cells with reasonable carrier mobilities because the electrons and holes exist everywhere in the bulk.

Our measurements showed that for these cells the photocurrent was linearly proportional to the incident light intensity, and that the shapes of the photocurrent vs voltage curves were independent of light intensity. These both indicate that bimolecular recombination was not an issue with these cells. Furthermore, we observed that the photocurrent scaled with the field inside the devices, which strongly indicates that geminate separation was the limiting factor in these cells. Increasing the field inside the device by applying an external bias extracted more of these charges. At high reverse bias, we extracted all of the ‘missing’ charges, indicating that we’ve accounted for all of the charges that were photogenerated in the active layer. This is a very important result since it shows that geminate recombination is the only major problem for these devices and that obtaining 100% internal quantum efficiency should be possible if this problem is fixed.

## **II. High Efficiency Solar Cells Using Triplet Excitons**

Triplet generating materials are attractive for use in solar cells because triplet excitons have long lifetimes and generally have large diffusion lengths. Most of these materials form triplets through intersystem crossing (ISC) from a singlet state through interactions with a heavy metal atom such as platinum or iridium. In pentacene, however, it has been demonstrated that triplets can form through an ultrafast fission process where a singlet exciton splits into a pair of triplets. If this process causes the majority of photoexcited singlets to become triplets, this would explain pentacene’s low fluorescence, large reported exciton diffusion length, and the high current obtained from pentacene based solar cells.

We took transient photocurrent measurements to determine if the time-dependence of the photocurrent matched the time-dependence of the triplet exciton population, implying that triplets were responsible for a significant fraction of the photocurrent generated. However, when we took the measurement, we observed a decay time slower than what we would expect from a singlet population but faster than what we would expect from a triplet population. We also measured transient photocurrent from a control device which was

made from a platinum porphyrin material (PtOEP) that is known to form triplets through fast intersystem crossing. Our (PtOEP) triplet control device showed the same behavior. We believed that this effect may have been due to interactions between PtOEP molecules in a concentrated film that are not present in the dilute films used commonly in the OLED community where this molecule was developed.

To verify that the triplet population was heavily influenced by the concentration of active molecules in the film, we measured the triplet phosphorescence spectrum as a function of concentration. We note that as the concentration of PtOEP was increased, the triplet phosphorescence signal decreased. Additionally, the spectrum became more and more red-shifted, suggesting that there was strong interaction between chromophores and possible aggregation of molecules.

We further investigated this by taking photoinduced absorption spectra of the PtOEP films to determine if there were triplets present that weren't contributing to the photocurrent for some reason. We noticed that in pure films, we could not see a triplet signal (the triplet signal was present in the dilute guest:host matrix). This observation is consistent with an observation made by Stefan Cook<sup>1</sup> who performed the same photoinduced absorption experiment on PCBM molecules. He notes that the triplet signal decreased and ultimately disappeared as he increased the concentration of PCBM in his sample. It appears that increasing the density of triplet-forming molecules in a film further mixes the singlet and triplet character of the excitons in the film. At low concentrations, the effect is to allow photogenerated singlets to transform into the lower energy triplets which have very long lifetimes. At high concentrations, the further mixing of the singlet and triplet state results in a lowering of the triplet lifetime from tens of microseconds to hundreds of nanoseconds. This faster decay is responsible for the shrinking of the triplet photoinduced absorption signal as well as the short lifetime observed in the transient photocurrent measurement.

We conclude that triplet excitons do not provide any significant advantage over singlet excitons in for several materials. While dilute guest:host configurations work well in OLEDs, where radiative recombination is desired and charge trapping is beneficial, solar cells require concentrated films. Because the triplet lifetime is shortened in such films, the triplet state is no longer advantageous.

Reference:

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### **III. Synthesis of Acenaphthyl and Phenanthrene Based Fused-aromatic Thienopyrazine Copolymers for Photovoltaic Applications**

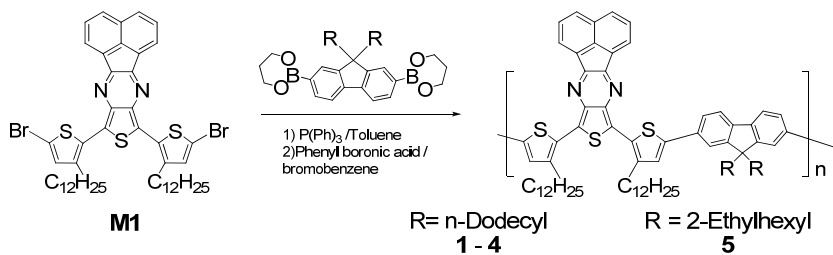
Semiconductive polymers may offer a route to photovoltaic and electronic function when light weight, flexibility, and low-temperature processing are required, as in bendable displays, solar panels, and smart textiles.<sup>1, 2</sup> Key material requirements are: i) high optical absorption at wavelengths  $> 500$  nm, where solar irradiance peaks;<sup>3</sup> ii) high field-effect mobility ( $\mu$ )  $> 0.1 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$ ;<sup>4, 5</sup> and iii) low-cost processing.<sup>6</sup> We report the performance of donor-acceptor copolymers designed seeking to achieve these properties.

A donor-acceptor structure reduces optical bandgap, enables absorption down to

the near infrared,<sup>7</sup> and may raise power conversion efficiency (PCE) in solar cells through increased photocurrent.<sup>8</sup> Benzothiadiazoles<sup>9-13</sup> and thienopyrazines<sup>14-21</sup> are commonly used as donors. Thienopyrazine (TP) allows modification of the pyrazine ring without introducing steric effects directly on the polymer backbone. Alkyl and aryl groups can be added at positions 2 and 3 to tune structural and electronic properties of the acceptor. In this work we introduced a rigid naphthalene moiety fused to the 2 and 3 TP positions. This gives the planar, electron rich  $\pi$ -face of acenaphtho[1,2-*b*]thieno[3,4-*e*]pyrazine (ACTP), which may promote  $\pi$ - $\pi$  stacking between polymer chains and lead to improved charge carrier mobility.

ACTP monomer **M1** was prepared from reaction of 3,4-diaminothiophene dihydrochloride and acenaphthenequinone, followed by bromination.<sup>22</sup> ACTP-Fluorene copolymers were made via Suzuki coupling (**Scheme 1**).<sup>23</sup> We synthesized five materials with different molecular weights (Mw) and side chains to study the effects of these variables on performance. The purified, dark-green powders were characterized by gel permeation chromatography (**Table 1**), differential scanning calorimetry, UV-vis absorption, and photoelectron spectroscopy, and have a low bandgap *ca.* 1.6 eV, with an absorption onset of *ca.* 800 nm.

We fabricated top-contact organic thin-film transistors (OTFT) from **1-5** to measure mobility. We drop-cast  $\sim$ 30 nm polymer films on heavily-doped n-type SiO<sub>2</sub>(300 nm)/Si wafers previously treated with octadecyltrimethoxysilane (OTS). Films were annealed at different temperatures, and gold electrodes were deposited through shadow masks. Annealing at 200 °C in N<sub>2</sub> yielded the best performance; lower temperatures gave lower mobilities, and higher temperatures caused film dewetting. Devices fabricated on phenyltrimethoxysilane treated surfaces did not dewet upon annealing, but their mobility was much lower.



**Scheme 1.** Preparation of ACTP-F copolymers.

Polymers **1-5** show *p*-channel transport, with mobility as high as  $0.2 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$ , of the same order of magnitude as the best polymer semiconductors reported.<sup>24-29</sup> The highest mobility was obtained with **1**. **Fig. 1** displays electrical characteristics of these OTFTs, under inert (**a, c**) and ambient conditions (**b, d**). There is no significant threshold voltage shift with changing environment, but mobility is higher in N<sub>2</sub> (**Fig 1. a, b**). The output traces (**Fig 1. c,d**) show somewhat late saturation and do not scale as expected in N<sub>2</sub>, but improve in air. This behavior was found in all polymers, and may be due to their relatively high HOMO ( $\sim$ -4.9eV) which makes them susceptible to oxygen doping ( $\sim$ -5.7 eV)<sup>18</sup> Transferring the devices back into N<sub>2</sub> recovers the same

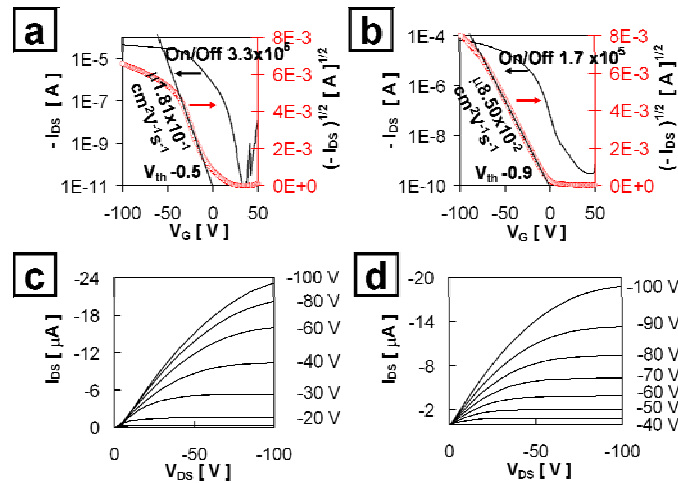
I-V characteristics. The average and max. mobility values of our materials are listed in **Table 1**.

**Table 1.** GPC and OTFT Characterization of ACTP-F Series.

ID	Mw <sup>a</sup>	PDI <sup>b</sup>	D <sup>c</sup>	$\mu$ Max. <sup>d</sup>	$\mu$ Ave. <sup>d</sup>	RSD <sup>e</sup>	I <sub>On</sub> /I <sub>Off</sub> <sup>f</sup>	V <sub>t</sub> <sup>g</sup>
<b>1</b>	10.0	1.6	8	0.195	0.166	15%	1.7E6	3.4
<b>2</b>	18.2	2.3	14	0.152	0.096	20%	7.9E6	6.8
<b>3</b>	20.7	2.2	16	0.115	0.077	36%	1.1E6	2.5
<b>4</b>	28.2	2.1	22	0.085	0.075	9%	1.2E6	10.9
<b>5</b>	11.9	1.5	10	0.016	0.009	42%	1.0E6	10.1

<sup>a</sup> Weight average molecular weight in kiloDaltons. <sup>b</sup> Polydispersity index. <sup>c</sup> Aprox. degree of polymerization. <sup>d</sup> Field effect mobility in cm<sup>2</sup>V<sup>-1</sup>s<sup>-1</sup> extracted from OTFT curves. <sup>e</sup> Relative standard dev. of the mobility. <sup>f</sup> Ratio of the on and off transistor currents. <sup>g</sup> Threshold bias in volts.

Mobility decreases with increasing Mw for copolymers **1-4**. This is likely a consequence of differences in film structure, and does not necessarily reflect intrinsic material properties.<sup>30</sup> We found by atomic force microscopy (AFM, **Fig. 2a-c**) that the grains of annealed films shrink and become less defined as Mw rises. **Fig. 2e** displays corresponding X-ray diffraction (XRD) spectra, showing that **1, 3** and **4** have equal d-spacing, but the intensity of this signal decreases with increasing Mw, indicating a lower degree of crystallinity in the higher Mw films. The 22.66 Å out of plane spacing in our films suggests partial interdigitation of side chains,<sup>31, 32</sup> favoring the formation of lamellar and  $\pi$ -stacked sheets and enhancing charge transport.<sup>33</sup> The inset shows slices of grazing incidence X-ray diffraction (GIXD) spectra from **1** and **4**. These show reduction in film ordering with increasing Mw, evidenced in the greater intensity of diffraction spots and the presence of in-plane diffraction rods for film **1** (white arrows), compared to the weak out-of-plane spot and diffraction arch on film **4**.

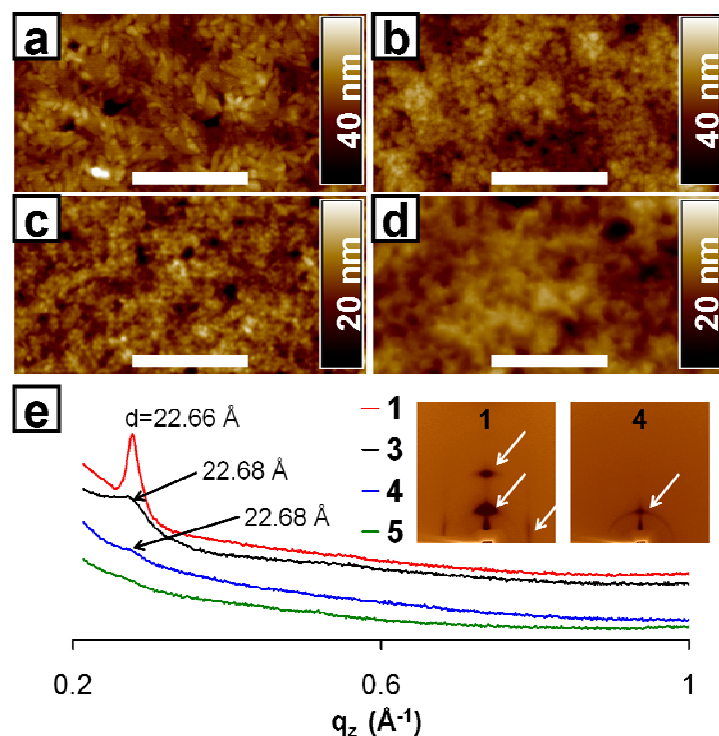


**Fig. 1** Representative I-V curves of OTFTs of **1**. (**a, c**) measurements performed in dry, inert environment. (**b, d**) same device tested in air.

The steric effects of the fluorene side chains largely impact mobility as seen in the case of **5**, with branched chains. This material has Mw just 10% higher than **1** but its mobility is twelve times smaller than that of **1**. We attribute such low mobility to decreased film order, manifested in the smoothed AFM of **Fig. 2d**, and in the featureless XRD in **Fig. 2e**.

We also evaluated **1**, **4** and **5** in bulk heterojunction solar cells made using indium-tin oxide (ITO) coated glass as substrate. First, 50 nm of poly(3,4-ethylenedioxythiophene)-poly(styrenesulfonate) were spin coated on the ITO, followed by spin-coating in N<sub>2</sub> environment mixtures of polymer and [6,6]-phenyl C<sub>61</sub>-butyric acid methyl ester (PC<sub>[61]</sub>BM) dissolved in orthodichlorobenzene. Films were solvent-annealed in a closed Petri dish,<sup>34</sup> and their typical thickness was ~110 nm. Aluminum electrodes were evaporated onto the polymer blend, and I-V characteristics were recorded in the dark and under simulated 1 sun AM 1.5G radiation. Using a 1:4 weight ratio of polymer:PC<sub>[61]</sub>BM we obtained the highest PCE for polymer **5** of 0.64%. Further optimization of cells from **5** by depositing *ca.* 1 nm LiF layer before the Al cathode to improve charge injection, resulting in a PCE of 1.38%. **Fig. 3** shows the I-V characteristics of this cell, and **Table 2** summarizes the solar cell performance of our materials.

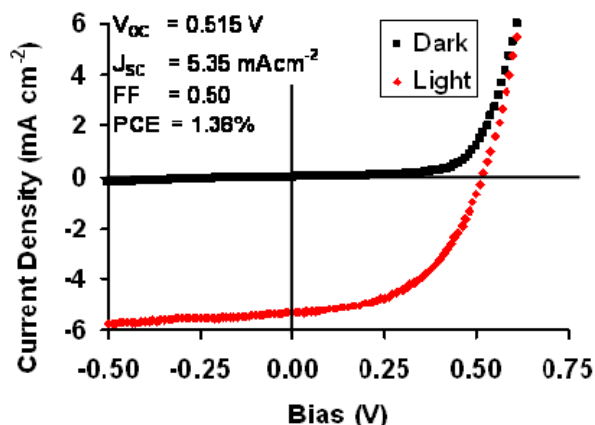
We also investigated the diode hole mobility ( $\mu_h$ ) in devices from **5** as a function of blending ratio by the space charge limited photocurrent method.<sup>35, 36</sup> We found  $\mu_h$  to be  $1.73 \times 10^{-6} \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$  in a 1:1 (by weight) blend of **5** PC<sub>[71]</sub>BM and  $1.34 \times 10^{-4} \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$  for a 1:4 blend. The  $\mu_h$  of the optimized blend of **5** compares well with the minimum  $\mu_h$  in P3HT:PC<sub>[61]</sub>BM 1:1 blends (optimal ratio for that system), which has been estimated to be  $\sim 3.4 \times 10^{-5} \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$ .<sup>37</sup> As noted in other bulk heterojunction systems,<sup>20, 35, 38</sup> we also observe that  $\mu_h$  of neat films of **5** ( $\sim 2.9 \times 10^{-6} \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$ ) is lower than that of the optimal blend, this difference being often attributed to PC<sub>[61]</sub>BM intercalation and enhancement in crystallinity in the blend. Despite its higher field effect mobility, polymer **1** had a lower PCE than **5**. PCE is influenced by several factors and we are investigating details of charge transport and morphology in our polymer:PCBM blends to gain a better understanding of these systems.



**Fig. 2.** Tapping mode AFM of annealed films of **1** (a), **3** (b), **4** (c) and **5** (d). A shows clearly defined grains, while **b-d** are less textured. (e) XRD spectra of the films. The gradual disappearance of the peak at  $\sim 22.66$  Å indicates disorder increases with Mw (**1-4**) or with incorporation of bulky side chains (**5**). Insets are GIXD slices of **1** and **4** showing the same decrease in film order.

ACTP-F copolymers may have limitations in solar cells due to low absorption coefficients ( $\alpha$ ). For example, films from **5** have  $\alpha \sim 1.6 \times 10^4 \text{ cm}^{-1}$  at 600 nm, compared to *ca.*  $1.8 \times 10^5 \text{ cm}^{-1}$  for P3HT.<sup>39</sup> By integrating the absorption spectrum of **5**:PC<sub>[61]</sub>BM blends (1:4 by weight), a theoretical maximum short-circuit current density of  $6.4 \text{ mAcm}^{-2}$  is obtainable, compared with the  $5.35 \text{ mAcm}^{-2}$  that we already achieved. This implies that the device performance is close to its potential, and is limited by absorption. We suspect the low absorption is due to weak coupling of the donor and acceptor, which may be improved by tuning the substituents and their locations.

In summary, we report new ACTP-F copolymers with a planar fused ring structure in the acceptor. These polymers showed high transistor mobility ( $\mu \sim 0.2 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ ) and moderate solar cell performance (PCE  $\sim 1.38\%$ ). The ACTP acceptor promotes lamellar crystalline structure when combined with linear alkyl chains in the fluorene and Mw  $\sim 10 \text{ kDa}$ . The crystallinity is highly dependent on the steric effects of the alkyl chains. Solar cell performance is promising, although fine-tuning to improve absorption coefficients is needed. All the above shows the many parameters available for tuning the performance of organic semiconductors in electronics and photovoltaics.



**Fig. 3** I-V characteristic for the solar cell made from **5**, 1:4 polymer:PC<sub>[61]</sub>BM by weight, optimized with an LiF/Al cathode.

**Table 2** Solar Cell Performance of the ACTP-F Series.

Polymer	% PCE	Fill Factor	V <sub>oc</sub> <sup>a</sup>	J <sub>sc</sub> <sup>b</sup>
<b>1</b>	0.39	0.40	0.435	2.27
<b>4</b>	0.49	0.59	0.485	1.71
<b>5</b>	0.64	0.32	0.485	4.10
<b>5 (LiF/Al)</b>	1.38	0.50	0.515	5.35

<sup>a</sup> Open-circuit voltage in volts. <sup>b</sup> Short-circuit current density in mAcm<sup>-2</sup>.

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- 1) R. Mondal, N. Miyaki, H.A. Becerril, J. Parmer, A. Mayer, M.L. Tang, M. McGehee, Z. Bao, **Chem. Mater.** submitted.
- 2) M.L. Tang, A.D. Reichardt, P. Wei, Z. Bao, **J. Am. Chem. Soc.** in press.
- 3) M.L. Tang, J.H. Oh, A.D. Reichardt, Z. Bao, **J. Am. Chem. Soc.** 131, 3733-3740, 2009.
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- 9) M. Tang, A. Reichardt, Z. Bao, **Adv. Func. Mater.**, 18, 1579-1585, 2008.
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