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Efficient Ternary Blend Bulk Heterojunction Solar Cells with Tunable Open-Circuit Voltage

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Supporting Information

ABSTRACT: To explore the potential of ternary blend bulk heterojunction (BHJ) photovoltaics as a general platform for increasing the attainable performance of organic solar cells, a model system based on poly(3-hexylthiophene) (P3HT) as the donor and two soluble fullerene acceptors, phenyl-C_{61}-butyric acid methyl ester ($PC_{61}BM$) and indene-C_{60} bisadduct (ICBA), was examined. In all of the solar cells, the overall ratio of polymer to fullerene was maintained at 1:1, while the composition of the fullerene component ($PC_{61}BM$:ICBA ratio) was varied. Photovoltaic devices showed high short-circuit current densities (J_{sc}) and fill factors (FF) (>0.57) at all fullerene ratios, while the open-circuit voltage (V_{oc}) was found to vary from 0.61 to 0.84 V as the fraction of ICBA was increased. These results indicate that the V_{oc} in ternary blend BHJ solar cells is not limited to the smallest V_{oc} of the corresponding binary blend solar cells but can be varied between the extreme V_{oc} values without significant effect on the J_{sc} or FF. By extension, this result suggests that ternary blends provide a potentially effective route toward maximizing the attainable $J_{sc}V_{oc}$ product (which is directly proportional to the solar cell efficiency) in BHJ solar cells and that with judicious selection of donor and acceptor components, solar cells with efficiencies exceeding the theoretical limits for binary blend solar cells could be possible without sacrificing the simplicity of a single active-layer processing step.

Bulk heterojunction (BHJ) solar cells based on a binary blend of a polymeric donor and a fullerene acceptor have seen rapid improvements in efficiency in recent years, from 2.5%[1] to ~8%.[2−6] However, the ultimate efficiency of such solar cells appears to be limited to 10−12%.[7−9] Ternary blend solar cells based on two donor components and one acceptor component (or one donor and two acceptors) have received far less attention but have been recognized as a potential route to increase the absorption breadth of a solar cell and consequently the short-circuit current density (J_{sc}).[10−16] Despite this potential advantage, it has been proposed that the open-circuit voltage (V_{oc}) of ternary blend solar cells would necessarily be pinned to the smaller V_{oc} of the corresponding binary blends of the constituent components, thus limiting the perceived impact of this device platform.[14] Here, using a model three-component system (Figure 1), we demonstrate for the first time that the V_{oc} of ternary blend BHJ solar cells is composition-dependent and can be tuned across the full range defined by the corresponding limiting binary blends without negatively impacting the fill factor (FF) or J_{sc} of the solar cells. The consequence is the possibility that with a judicious choice of components, the attainable $J_{sc}V_{oc}$ product (and by extension the efficiency $\eta = (J_{sc} \cdot V_{oc} \cdot \mathrm{FF})/P_{in}$, where P_{in} is the intensity of the incident light) in a single-layer ternary blend solar cell could be higher than is achievable with a standard binary blend solar cell.

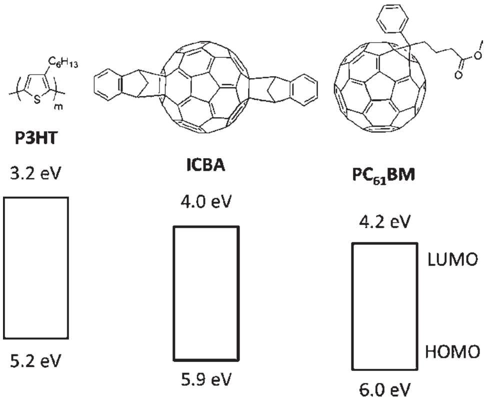

Figure 1. Structures and corresponding HOMO and LUMO energy levels of P3HT, ICBA, and $PC_{61}BM$.

In a binary blend BHJ polymer−fullerene solar cell, the J_{sc} is proportional to the product of the absorption breadth and absorption intensity of the active layer, which is generally limited by the band gap (E_g) of the donor polymer.[17] In principle, a smaller E_g gives a broader wavelength range of light absorption and consequently a higher J_{sc}. Conversely, the V_{oc} cannot exceed the energetic difference between the donor HOMO ($HOMO_D$) and the acceptor LUMO ($LUMO_A$).[7,18] Considering the fact that an energetic driving force for charge transfer must exist (approximated by the $LUMO_D - LUMO_A$ offset[19]), a high J_{sc} is favored by a shallow $HOMO_D$ and a high V_{oc} by a deep $HOMO_D$. Ultimate optimization is found through adjusting the frontier orbital energies of the donor and acceptor components to balance the opposing quantities of J_{sc} and V_{oc}, targeting a maximum value of the $J_{sc}V_{oc}$ product rather than a maximum attainable value for either J_{sc} or V_{oc}. As a consequence, the ultimate efficiency of binary blend polymer−fullerene BHJ solar cells is limited to 10−12%.

As a route toward higher efficiency, tandem solar cells, in which two (or more) subcells absorbing light in different regions

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of the solar spectrum are connected either in series or parallel, have become an increasingly explored alternative to simple BHJ solar cells.[20,21] The ultimate achievable efficiency of a tandem cell with two absorbing layers is predicted to be 14−15%.[8,22] In the case of serial connection of the subcells, the V_{oc} of the tandem device can approach the sum of the V_{oc} values of the individual subcells[23] while the J_{sc} can at best approach the highest J_{sc} of the subcells.[20,23,24] On the other hand, with parallel connection of the subcells, the J_{sc} approaches the sum of those for the individual subcells[20] but the V_{oc} is limited to the minimum V_{oc} of the individual subcells.[20] Thus, it is clear that tandem cells also do not allow independent and concurrent optimization of both J_{sc} and V_{oc} beyond that of the individual subcells. Another drawback of tandem solar cells is the increased complexity of cell design and fabrication,[20,23] which is in contrast to the attractive simplicity of the single-step solution processing of the active layer in BHJ solar cells.[7]

Ternary blend BHJ solar cells offer a distinct platform and an alternative approach for increasing the attainable $J_{sc}V_{oc}$ product while retaining the simplicity of a single active-layer processing step. There is a growing body of literature describing a number of variants of the ternary blend system based on either two polymer donors and a fullerene acceptor,[10,12−16] one polymer donor and two acceptors,[25−29] a polymer donor, a fullerene acceptor, and a small molecule/dye,[11,30−38] or a polymer donor with a nanoparticle and a fullerene as acceptor.[39−43] In many cases, an increase in the breadth of the spectral response of the ternary blends has been observed relative to the corresponding limiting binary blends, often leading to a larger J_{sc}.[10−16] Conversely, the V_{oc} is proposed to be pinned to the smallest V_{oc} of corresponding binary blends.[14,16,44] To this end, it is thought that a limiting $HOMO_D−LUMO_A$ interaction controls the V_{oc}, since dominant hole transport and collection occurs through the donor component with the highest-lying HOMO (and analogously, electron transport and collection through the lowest-lying LUMO), independent of the origin of photocurrent generation.[14] However, in a limited number of cases, the V_{oc} seems to be tunable in the three-component system, although at the expense of a marked and steady decrease in the FF as the amount of the third component increases.[25,30,31] Nonetheless, these isolated observations of composition-tunable V_{oc} suggest that both J_{sc} and V_{oc} are composition-dependent in ternary blend BHJ solar cells and that neither value is necessarily limited to the lesser quantity of the corresponding binary blend solar cells. In support of this, here we communicate for the first time an example of a ternary blend BHJ solar cell in which the V_{oc} is tunable across the full composition range of the components while a high FF is maintained at all compositions.

The model ternary system containing poly(3-hexylthiophene) (P3HT) as the donor and phenyl-C_{61}-butyric acid methyl ester ($PC_{61}BM$) and indene-C_{60} bisadduct (ICBA) as acceptors used here (Figure 1) was chosen for several reasons. First is the similarity in the chemical structures of the acceptors, which is envisioned to give good miscibility between them. Second is the excellent miscibility known in the limiting binary polymer:fullerene blends, resulting in similar polymer−fullerene ratios (close to 1:1) and processing conditions (post-aluminum annealing at 150 °C) necessary for the optimal solar cell performance.[45,46] Moreover, high efficiencies with FF > 0.6 have been observed in both binary blend BHJ solar cells. Finally, the two limiting polymer−fullerene binary blends give significantly different values of V_{oc} (0.6 V for P3HT:$PC_{61}BM$[45] and 0.84 V for P3HT:ICBA[46]) because of the different positions of the acceptor LUMOs,[7] as shown in Figure 1.

Table 1. Photovoltaic Properties of P3HT:PC_{61}BM:ICBA Ternary Blend BHJ Solar Cells at Different Fullerene Ratios[a]

P3HT:PC_{61}BM:ICBA	J_{sc} (mA/cm^2)	V_{oc} (V)[b]	FF	η (%)
1:1:0[c]	9.90	0.605	0.60	3.57
1:0.9:0.1[d]	9.22	0.618	0.59	3.29
1:0.8:0.2[d]	9.11	0.631	0.57	3.28
1:0.7:0.3[e]	8.58	0.649	0.58	3.22
1:0.6:0.4[f]	8.31	0.669	0.58	3.11
1:0.5:0.5[g]	8.27	0.688	0.57	3.18
1:0.4:0.6[e]	8.18	0.709	0.57	3.22
1:0.3:0.7[h]	8.14	0.741	0.57	3.34
1:0.2:0.8[d]	8.19	0.769	0.59	3.69
1:0.1:0.9[d]	8.18	0.804	0.60	3.91
1:0:1[h]	8.23	0.844	0.58	3.98

[a] Devices were spin-coated from CB and after aluminum deposition were annealed at 150 °C under N_2 for the specified times. [b] Standard deviations of less than 0.005 were observed in all cases for averages over eight pixels. [c] 60 min. [d] 20 min. [e] 40 min. [f] 30 min. [g] 50 min. [h] 10 min.

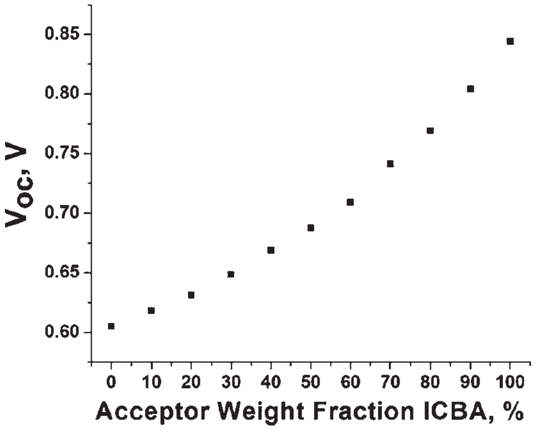

Figure 2. V_{oc} for the ternary blend BHJ solar cells as a function of the amount of ICBA in the blend.

Photovoltaic devices containing ternary blends in the conventional device configuration ITO/PEDOT:PSS/P3HT:PC_{61}BM:ICBA/Al were fabricated in air. In order to compare the device parameters, all of the optimized devices had active-layer thicknesses of 95−105 nm. Additionally, the concentration of all polymer−fullerene solutions was kept constant (10 mg/mL in P3HT), as was the annealing temperature (150 °C). Furthermore, in all cases the overall P3HT:fullerene weight ratio was maintained at 1:1. Table 1 lists the average values of J_{sc}, V_{oc}, FF, and η obtained under simulated AM 1.5G illumination (100 mW/cm^2) as the PC_{61}BM:ICBA ratio was varied.

Several significant observations can be made from the data in Table 1. Importantly, as is also illustrated in Figure 2, V_{oc} of the three-component solar cells showed a continuous increase from 0.605 to 0.844 V as the amount of ICBA in the ternary blend increased. This establishes that in ternary blend BHJ solar cells, V_{oc} is not necessarily pinned to the smallest V_{oc} of the corresponding binary blends. Table 1 also shows that high FF values (>0.57) were observed for all of the photovoltaic devices, independent of the fullerene ratio. This can be attributed to balanced, trap-free charge

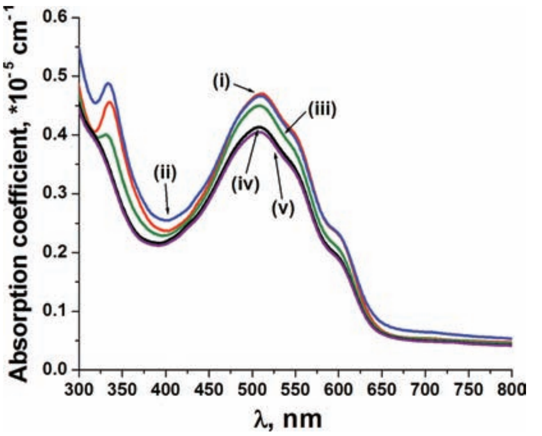

Figure 3. UV−vis absorption spectra of thin films spin-coated from CB and annealed at 150 °C under N_2 for 20 min with P3HT:PC_{61}BM:ICBA ratios of (i) 1:1:0 (red), (ii) 1:0.8:0.2 (blue), (iii) 1:0.5:0.5 (green), (iv) 1:0.2:0.8 (black), and (v) 1:0:1 (purple).

transport through the bulk[47] and favorable morphology.[48] Transmission electron microscopy (TEM) [see the Supporting Information (SI)] showed very similar, bicontinuous blends with nanometer-scale phase separation, independent of the fullerene ratio. Thus, charge separation and transport do not appear to be hindered in the ternary blend solar cells.

In contrast to the V_{oc} trend observed in Table 1, J_{sc} was found to decrease with increasing ICBA content. This observation was explained using external quantum efficiency (EQE) measurements (see the SI) and comparison with the absorption coefficients of the various blend films. The highest photocurrent response was observed for the P3HT:PC_{61}BM blend, while introduction of ICBA into the blends led to a gradual reduction in the photocurrent intensity, with the minimum value reached for P3HT:ICBA solar cells. In order to investigate further the origin of the decrease in EQE at high ICBA contents, the optical properties of annealed P3HT:PC_{61}BM:ICBA blends at various fullerene ratios in thin films spin-coated from chlorobenzene (CB) were studied using UV−vis spectroscopy, as shown in Figure 3. The introduction of ICBA resulted in a decrease in the absorption coefficient of the thin film that was more pronounced as the amount of ICBA in the blend increased. This decrease can be explained using the absorption coefficients and profiles of PC_{61}BM and ICBA (see the SI). For ICBA, the absorption strength in the visible is significantly less that that for PC_{61}BM. Since all the devices from Table 1 have the same active-layer thickness, the introduction of ICBA into the film decreases the number of photons absorbed, thus leading to a decrease in EQE and by extension J_{sc}.

To verify the possibility of achieving higher J_{sc} in the case of high ICBA content, P3HT:PC_{61}BM:ICBA ternary blend solar cells at 1:0.5:0.5 and 1:0:1 ratios were optimized (see the SI) to film thicknesses of 137 and 174 nm, respectively. These optimized photovoltaic devices showed improved J_{sc}, FF, and η values of 9.82 mA/cm^2, 0.59, 3.92% and 9.23 mA/cm^2, 0.59, 4.55%, respectively, with essentially no change in V_{oc} (0.682 and 0.839 V, respectively) relative to the devices reported in Table 1. Thus, high J_{sc} and FF are possible in ternary blend solar cells, independent of the ratio of the three components in the blend and without effect on the compositional dependence of V_{oc}.

Another important feature of the thin-film absorption spectra in Figure 3 is the presence of the strong vibronic feature near 600 nm for all of the P3HT:PC_{61}BM:ICBA blends. This shoulder is common for P3HT thin films[49] and generally is ascribed to the interchain vibrational absorption induced by a high degree of ordering and strong interchain interaction.[50] To study the effect of ternary blends on the degree of P3HT crystallinity, grazing-incidence X-ray diffraction (GIXRD) was used (see the SI). For all of the films, peaks corresponding to the interchain distance (100) for P3HT were observed in the range 16.4−16.7 Å with similar intensities, indicating that the ability to obtain semicrystallinity in P3HT is not hindered in the ternary blends.

Beyond this model ternary system, which has a fixed spectral range for the two limiting binary blends, it is clear that the composition-tunable V_{oc} in ternary blends offers the potential for higher efficiencies than are attainable in binary blends if simultaneous tuning of J_{sc} is also targeted. This can be illustrated by considering a case of two hypothetical donor polymers (D1 and D2) and a fullerene acceptor (A), where $E_{gD1} > E_{gD2}$ and $HOMO_{D1}$ is lower than $HOMO_{D2}$. In this case, it is assumed that the D1:A binary blend displays $V_{oc} = 0.8$ V, $J_{sc} = 8.0$ mA/cm^2, and FF = 0.6, giving an efficiency of 3.8%, and the D2:A binary blend shows $V_{oc} = 0.5$ V, $J_{sc} = 14.0$ mA/cm^2, and FF = 0.6, giving an efficiency of 4.2%. Combining D1, D2, and A in a ternary blend could lead to higher J_{sc} for the ternary blend than for either limiting binary blend as a result of more uniform spectral coverage of absorption (as has been demonstrated previously[10−16]). Even a modest increase in J_{sc} to 15.0 mA/cm^2 could couple with an intermediate V_{oc} of 0.65 V to give an efficiency of 5.9% at FF = 0.6. The ternary blend could thus give a higher efficiency than either limiting binary blend as a result of the higher attainable $J_{sc}V_{oc}$ product. On the basis of the tunability of V_{oc} established here, it is proposed that with a judicious choice of components, ternary blends with efficiencies exceeding the 10−12% maximum predicted for binary blends could be achieved. Future work will focus on broadening the scope of investigated ternary systems beyond the illustrative model system studied in this work.

In summary, we have fabricated ternary blend BHJ solar cells containing P3HT as the donor and two soluble fullerenes, PC_{61}BM and ICBA, as acceptors. Devices were tested at different acceptor ratios and showed uniformly high J_{sc} and FF. Importantly, the V_{oc} of the three-component solar cells could be tuned between the limiting V_{oc} values of the corresponding binary blend solar cells. Taken together with previous literature, these results now establish that both V_{oc} and the spectral response (J_{sc}) of ternary blend solar cells are composition-dependent. This suggests that J_{sc} and V_{oc} are not necessarily constrained by the same factors and with the same limitations in ternary blends as they are in binary blends. These results indicate that ternary blend BHJ solar cells are promising candidates for the next generation of solution-processable solar cells and have the potential to overcome the predicted ultimate efficiencies for binary blend photovoltaic devices, as they can exhibit potentially higher attainable $J_{sc}V_{oc}$ products through judicious component selection and compositional control without sacrificing the attractive simplicity of processing in single-active-layer BHJ solar cells.

■ ASSOCIATED CONTENT

Ⓢ Supporting Information. Synthetic and solar cell fabrication procedures and NMR, GIXRD, UV−vis, TEM, EQE, and $J-V$ data. This material is available free of charge via the Internet at http://pubs.acs.org.

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