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14. ABSTRACT The focus of this research project was to develop novel charge selective materials to improve the performance of polymer solar cells. Interfacial engineering has been identified as an important section in bulk heterojunction polymer solar cells to maximize power conversion efficiency since it can effectively alleviate energy barriers at interfaces in multi-layered architectures, particularly for the organic/electrode interfaces, and thus to facilitate charge transport/extraction in the device. In this regard, a series of self-doped fullerene materials with much increased conductivity compared to the parent semiconducting fullerenes was developed. The self-doping mechanism was clarified to result from the anion-induced electron transfer (AIET) from constituent iodide to adjacent fullerene core during film evolution. Benefitting from their proper energy levels and decent conductivity, these fullerene derivatives can serve as efficient electron extraction layers (EELs) in both conventional and inverted PSCs. On one hand, they serve as the cathode-independent interlayers in the conventional PSCs owing to their electrode WF tuning capability. On the other hand, their high conductivities enable them to be less thickness sensitive (16-50 nm) in the inverted structure to result in a promising PCEs of up to 9.62% in the inverted PTB7-Th:PC ₇₁ BM device. Moreover, extending from this n-doping strategy, further development resulted in a crosslinkable self-doped fullerene composite interlayer, which possesses respectable solvent resistance, decent conductivity, and electrode WF tuning capability. In addition, these fullerene-based EELs have also been applied into perovskite solar cells (PVSCs) to obtain improved device performance and stability. All these results affirm the great potential of using FPI-based EELs for achieving high-efficiency PSCs and PVSCs.						
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Final Report for AOARD (FA2386-11-1-4072)

“Development of Efficient Charge-Selective Materials for Bulk Heterojunction Polymer Solar Cells”

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Abstract:

The focus of our AOARD sponsored project is to develop novel charge selective materials to improve the performance of polymer solar cells. During this investigation, we have been collaborating with Professor Kung-Hwa Wei (NCTU) through information and material exchanges to expedite the progress. Interfacial engineering has been identified, as an important section in bulk heterojunction polymer solar cells to maximize the power conversion efficiency since it can effectively alleviate the energy barriers existed at the interfaces in the multi-layered architecture, particular for the organic/electrode interfaces, to facilitate the charge transport/extraction in the device. In this regard, we have first developed a series of self-doped fullerene materials with much increased conductivity compared to the parent semiconducting fullerenes. The self-doping mechanism was clarified to result from the anion-induced electron transfer (AIET) from constituent iodide to adjacent fullerene core during film evolution. Benefitting from their proper energy levels and decent conductivity, these fullerene derivatives can serve as efficient electron extraction layers (EELs) in both conventional and inverted PSCs. On one hand, they serve as the cathode-independent interlayers in the conventional PSCs owing to their electrode WF tuning capability. On the other hand, their high conductivities enable them to be less thickness sensitive (16-50 nm) in the inverted structure to result in a promising PCEs of up to 9.62% in the inverted PTB7-Th:PC₇₁BM device. Moreover, extending from this n-doping strategy, we have further developed a crosslinkable self-doped fullerene composite interlayer, which possesses respectable solvent resistance, decent conductivity, and electrode WF tuning capability. In addition, we have also applied these fullerene-based EELs into the perovskite solar cells (PVSCs) to obtain improved device performance and stability. All these results affirm the great potential of using FPI-based EELs for achieving high-efficiency PSCs and PVSCs.

Introduction:

Polymer solar cells (PSCs) have attracted considerable research interest in the past decade due to their great advantages of low-cost, light-weight, and decent flexibility.¹ Compared to the conventional inorganic counterparts, PSCs are highly compatible to the roll-to-roll process for massive production. At present, the bulk-heterojunction (BHJ) PSC is the most widely investigated type, in which the BHJ active layer is based on a blend of a conjugated polymer as an electron donor (D) and a fullerene derivative as an electron acceptor (A).^{2, 3} Such phase-separated blend can provide sufficient D-A interfaces in photoactive layer to

dissociate the excitons and transport the free charge carriers to the collecting electrodes simultaneously.

The typical device configuration of a BHJ PSC comprises an organic photoactive layer sandwiched between two charge-collecting electrodes. In this respect, interfacial engineering becomes very important to maximize the power conversion efficiency (PCE) of PSC since it can effectively alleviate the energy barriers existed at the interfaces, particular for the BHJ/electrode interfaces, to facilitate the charge transport/extraction in the device.^{4, 5} Basically, the holes will be collected at anode and electrons will be extracted at cathode due to the built-in electrical field created by the distinct work functions (WF) of the asymmetric electrodes as well as the kinetic selectivity of electron-hole pairs in BHJ layer. Accordingly, the nature of electrical contact of the organic/electrode interfaces will have a huge influence on the resulting photovoltaic performance including open-circuit voltage (V_{OC}), short-circuit current (J_{SC}), and fill factor (FF).^{4, 6-10}

In principle, there are several important requirements need to be considered for the design of an ideal interfacial material. First, the interfacial materials should have the capability to tune the WF of electrode for Ohmic contact between the electrodes and photoactive layers. For example, various interfacial materials have proven to introduce the interfacial dipole to enable the energy level alignment at the organic/electrode interfaces. Second, they should have appropriate energy levels along with decent electrical conductivity for efficient charge transport/extraction. The suitable energy levels of interlayers can improve the electrode selectivity to extract only one type carriers while blocking the other type carriers. Third, high transparency of these interfacial materials is required, which can prevent the parasitic absorption loss of photoactive layer to reduce the optical loss in the device. Fourth, they should possess reasonable robustness to realize the construction of mutli-layered devices. Besides the abovementioned requirement, other factors such as facile preparation, proper surface energy for the optimized morphology evolution of BHJ layers, or improving the adhesion of each constituent interlayers in the stratified devices are sometimes also needed to be taken into account.

As stated in the original proposal, we aim to develop the novel charge-selective interfacial materials to enhance the device performance of PSC. Regarding the mentioned criteria for interfacial materials, we have successfully developed a series of highly efficient fullerene-based interfacial materials for facilitate the electron collection in the PSCs. Inspired by the success of p-type PEDOT:PSS, we have first synthesized a series of self-doped fullerene materials with much increased conductivity compared to the parent semiconducting fullerenes. Benefitting from their proper energy levels and decent conductivity, these fullerene derivatives can serve as efficient electron extraction layers in both conventional and inverted PSCs. On one hand, owing to their electrode WF tuning capability, they also serve as the cathode-independent interlayers in the conventional PSCs. On the other hand, their high conductivity enable them to have less thickness sensitivity (16-50 nm) in the inverted structure and can result in a promising PCEs up to 9.62% in the inverted PTB7-Th:PC₇₁BM device. Besides, we have also applied these fullerene-based EELs into the perovskite solar cells (PVSCs) to obtain improved device performance and stability.

Results and Discussion:

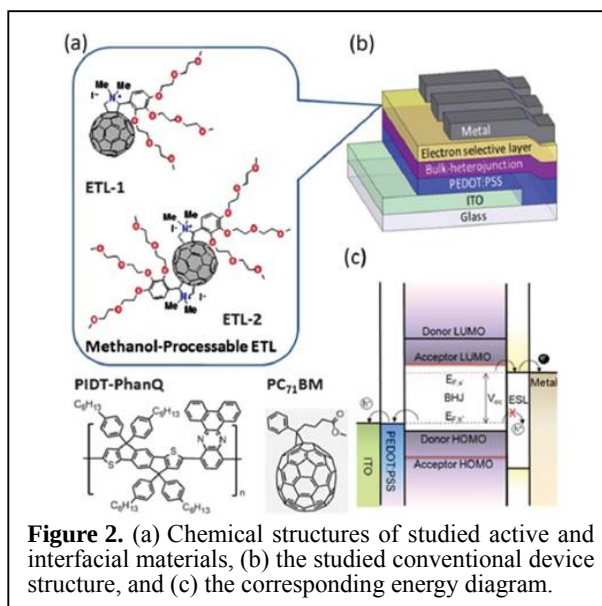
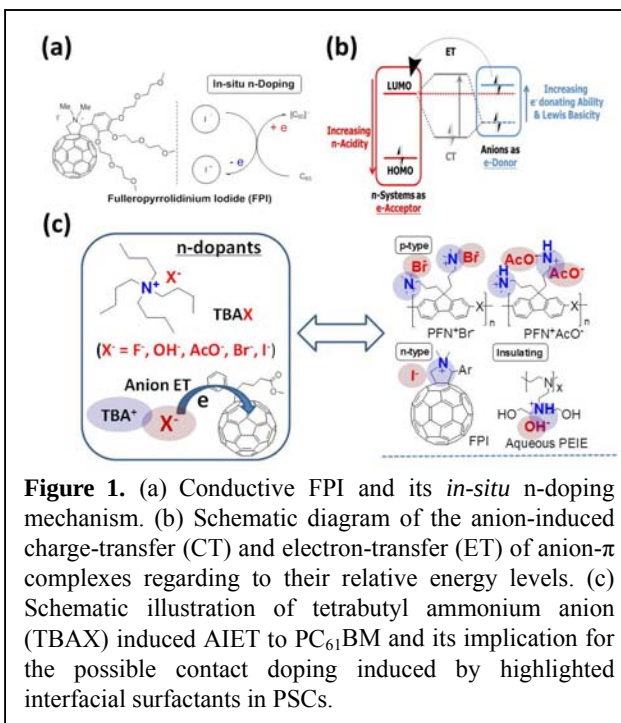
(1) Development of efficient fullerene-based electron extraction layer (EELs) with self-doped property and their application in PSCs for high performance

We have first described a series of solution-processable conductive fullerenes, fulleropyrrolidinium iodides (FPIs, **Figure 1a**), which show a decent conductivity of 2×10^{-2} S/cm. The underlying reason for such high conductivity of FPIs compared to other neutral fullerene derivatives was unveiled to arise from the anion-induced electron transfer (AIET) from constituent iodide to adjacent fullerene core during film evolution (**Figure 1b-c**).^{11, 12} As illustrated in **Figure 1b**, due to the anion- π interaction, the fullerene π -systems with electron-accepting abilities can interact with the approaching anions serving as Lewis base to form anion- π complexes.¹³ Consequently, these complexes will behave like D-A charge-transfer (CT) complexes to exhibit CT or electron-transfer (ET) characteristics, where the anion acts as a donor and the π -system acts as an acceptor. Notably,

the advantage of such AIET in doping is its facile solution-processability at room temperature under room light without the need to engage any harsh thermal/photo-activation process.

FPI can also serve as a dopant to dope PC₆₁BM to achieve high conductivity (3.2×10^{-2} S/cm) in thin film as well. Thereby, the interfacial doping of the nearby PCBM in BHJ layer is also expectable. Such conductive interface can be speculated to create barrier-less contact for better electron transport/extraction and enhance device performance.^{14, 15} Besides, owing to its zwitterion characteristic, FPI can also lower the WF of electrode to enable a better energy level alignment at the BHJ/electrode interface. This function also enables the utilization of stable metals such as Ag as the top electrodes, providing the device with respectable environmental stability.

Based on these promising advantages, the device performance of PSCs using the FPI-based EELs was demonstrated to be largely enhanced both in the conventional^{14, 15} and inverted structures¹⁶. Shown in **Figure 2** is the illustration of the FPI-based EELs employed in the conventional PIDT-PhanQ:PC₇₁BM BHJ



devices. Owing to their decent conductivity, appropriate energy levels, and electrode WF tuning capability, the FPI-based EELs can greatly improve the device performance, especially for the V_{OC} , since FPIs enable the energy level alignment at the organic/cathode interface. Note that the large increase in device V_{OC} is independent of the choice of cathode metal due to pinning of the metal E_F to that of FPIs upon equilibration. Mott-Schottky (MS) analysis of the BHJ/cathode interfaces yields a built-in potential defined by the difference between the Fermi level of the BHJ E_F^p and the effective cathode WF Φ_{cathode} . The observed changes in V_{BI} are reflected in the magnitude of the change in V_{OC} , as presented in **Figure 3**.

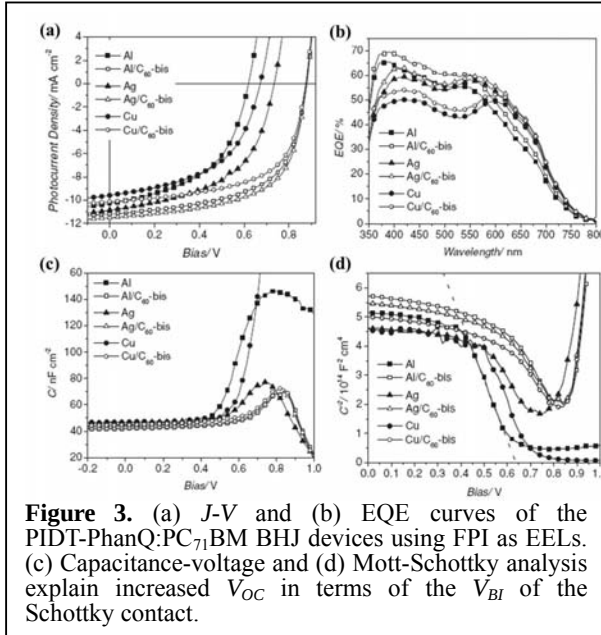


Figure 3. (a) J - V and (b) EQE curves of the PIDT-PhanQ:PC₇₁BM BHJ devices using FPI as EELs. (c) Capacitance-voltage and (d) Mott-Schottky analysis explain increased V_{OC} in terms of the V_{BI} of the Schottky contact.

charge extraction (CE) measurements clearly revealed that the employment of this EEL can largely reduce the nongeminate recombination in the device, as shown in **Figure 5**, due to its decent electron transporting property. As a result, a promising PCE up to 9.62% could be achieved in the inverted PTB7-Th:PC₇₁BM device (**Figure 4c**).

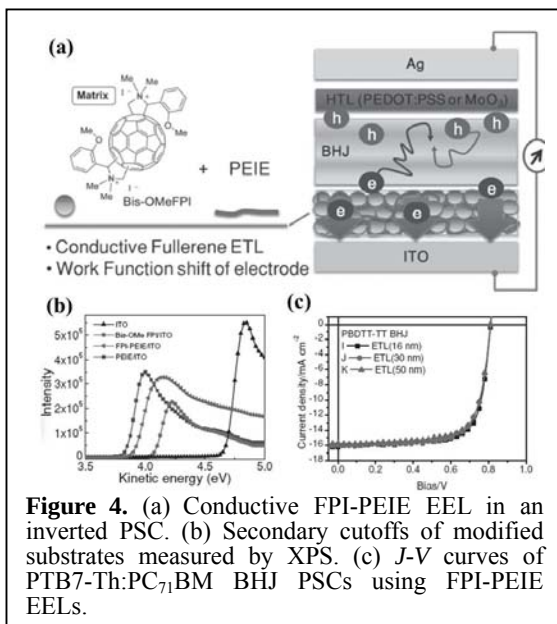


Figure 4. (a) Conductive FPI-PEIE EEL in an inverted PSC. (b) Secondary cutoffs of modified substrates measured by XPS. (c) J - V curves of PTB7-Th:PC₇₁BM BHJ PSCs using FPI-PEIE EELs.

Following these successful achievement of conductive fullerenes, we further developed an fullerene-based composite consisting of a conductive FPI derivative (Bis-OMe) and small amount of insulating PEIE to serve an excellent EEL in the inverted PSC (**Figure 4a**).¹⁶ On one hand, such hybrid EEL can tune the WF of electrode (**Figure 4b**) to promote the Ohmic contact at the BHJ/electrode interface. On the other hand, inheriting the self-doping property from Bis-OMe, this EEL possesses decent conductivity and can introduce the interfacial contact doping of the PCBM in BHJ as mentioned earlier, which enables itself to function well in thicker film (~50 nm) (**Figure 4c**). Also, the transient photovoltage (TPV) and

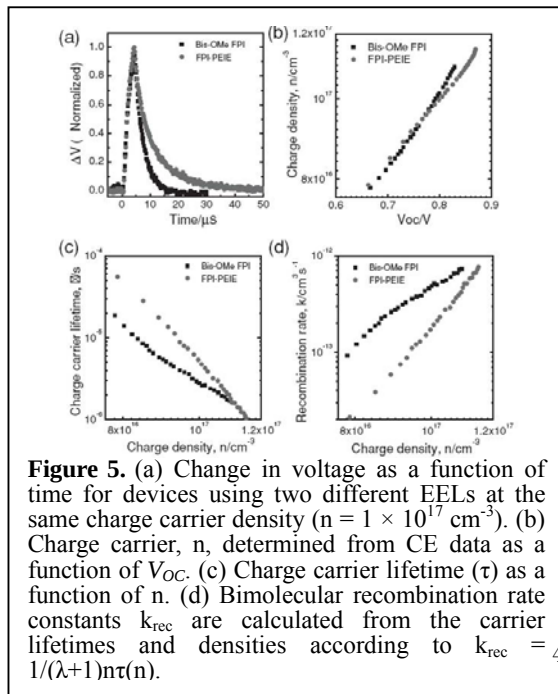


Figure 5. (a) Change in voltage as a function of time for devices using two different EELs at the same charge carrier density ($n = 1 \times 10^{17} \text{ cm}^{-3}$). (b) Charge carrier, n , determined from CE data as a function of V_{OC} . (c) Charge carrier lifetime (τ) as a function of n . (d) Bimolecular recombination rate constants k_{rec} are calculated from the carrier lifetimes and densities according to $k_{\text{rec}} = 4 / (\lambda + 1) n \tau(n)$.

(2) Development of cross-linkable fullerene-based EELs with decent conductivity and solvent resistance for efficient PSCs

Another important task of this proposal is to develop a charge-transporting interlayer with respectable solvent resistance. In this regard, we developed an *in-situ* doping and crosslinking fullerene-based EEL by blending a self-doped fullerene as a dopant with a thermally crosslinkable fullerene matrix (**Figure 6a**).¹⁷ It is worthwhile to note that the *in-situ* doping and crosslinking of this composite can be simply processed without engaging any harsh treatment. Owing to the decent conductivity introduced by bis-FPI and solvent resistance brought by the crosslinkable fullerene matrix, the PCE of the PIDT-PhanQ:PC₇₁BM BHJ device using this EEL can be significantly improved to 5.26% from 2.43% (the device using the EEL without incorporating bis-FPI dopants (**Figure 6b**)). The importance of introducing conductive bis-FPI is also evidenced by comparing the device using neutral bis-FP as dopant. As shown in **Figure 6b**, it clearly manifested that *in-situ* doping in the EEL can largely improve the overall device performance.

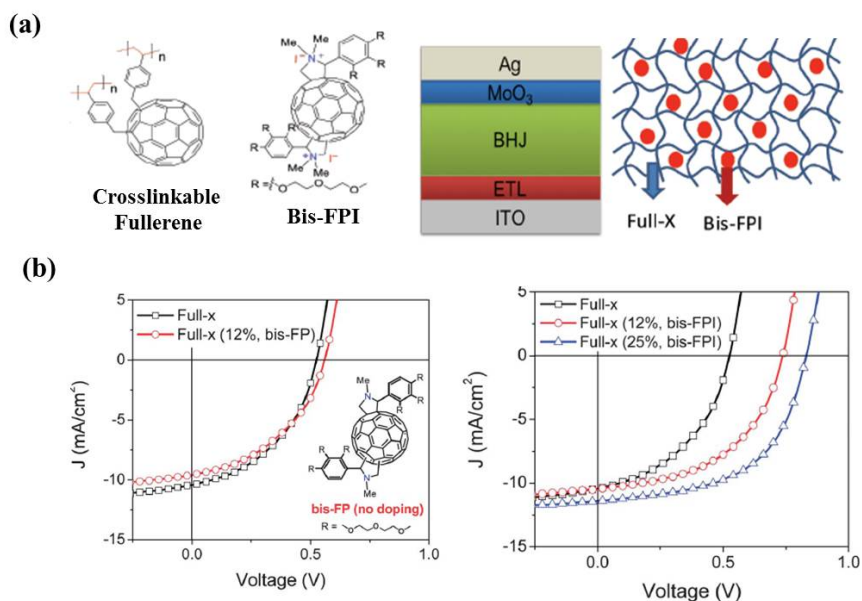


Figure 6. (a) Chemical structures of the crosslinkable fullerene and bis-FPI and the device configuration. (b) *J*-*V* characteristics of PCs fabricated using Full-x with different concentrations of bis-FPI and bis-FP.

(3) Application of fullerene-based EELs in perovskite solar cells (PVSCs)

As mentioned, the FPI-based EELs are cathode-independent interlayers, which can allow the usage of stable metals such as Ag as the top electrodes to enhance the device's environmental stability simultaneously. For organometal halide perovskite solar cells (PVSCs), this becomes an even

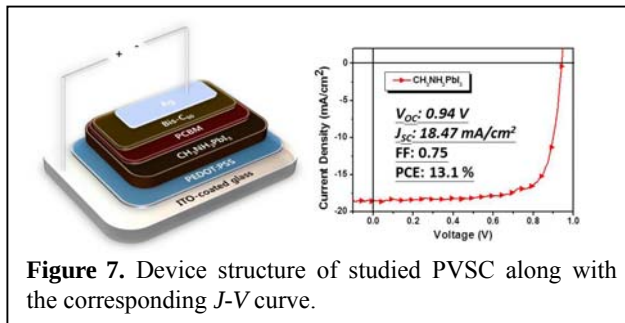


Figure 7. Device structure of studied PVSC along with the corresponding *J*-*V* curve.

important issue since the organometal halide perovskite itself is very sensitive to the humidity. Accordingly, the derived device will have a very short lifetime if the electrode is also not stable. In this regard, to employ a stable top electrode not only can increase the device reliability but also can impede the permeation of oxygen and humidity into perovskite to extend the device's lifetime. We recently successfully applied the FPI EEL into the planar heterojunction (PHJ) PVSC to afford high-performance and stability by constructing the conventional p-i-n PVSC with the configuration of ITO/PEDOT:PSS/perovskite/PC₆₁BM/FPI/Ag (**Figure 7**).¹⁸⁻²⁰ We demonstrated that the FPI can facilitate the charge transport at the PC₆₁BM/cathode interface while allowing the utilization of Ag as top, stable electrode due to its WF tuning capability. As a result, a preliminary PCE of 13% can be obtained in the PVSCs using FPI EELs.

All these results affirm the great potential of using FPI-based EELs for achieving high-efficiency PSCs. We believe the combined highly conductive and robust ETL will establish a very solid material foundation for enabling the fabrication of multi-junction PSCs and PVSCs to reach high efficiency, low-cost, and good stability. By applying these tailored interfacial materials to PSCs and PVSCs, it will bring us closer understanding of the connections between molecular structures and performance that are urgently needed to reach theoretical efficiency limits.

Through this AOARD support, we have published more than 40 journal articles and made very significant impact on improving the performance of solution processible (both OPV and perovskite-based) solar cells. Our Taiwanese partner, Prof. Kung-Hwa Wei's group has also been benefitted by the concept of using interfacial engineering to improve the device performance using their own materials. We are in the process of putting several joint papers together for publications.

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