High Performance Tandem Organic Solar Cells using HSolar as the Interconnecting Layer

Carr Hoi Yi Ho, Taesoo Kim, Yuan Xiong, Yuliar Firdaus, Xueping Yi, Qi Dong, Jeromy J. Rech, Abay Gadisa, Ronald Booth, Brendan T. O'Connor, Aram Amassian, Harald Ade, Wei You, Thomas D. Anthopoulos, Franky So**

Dr. C. H. Y. Ho, Dr. T. Kim, Dr. X. Yi, Q. Dong, Prof. A. Amassian, Prof. F. So
Department of Materials Science and Engineering, and Organic and Carbon Electronics Lab (ORaCEL)
North Carolina State University
Raleigh, NC 27695, USA
Email: hho3@ncsu.edu; fso@ncsu.edu

Y. Xiong, Dr. A. Gadisa, Prof. H. Ade
Department of Physics, and Organic and Carbon Electronics Lab (ORaCEL)
North Carolina State University
Raleigh, NC 27695, USA

Dr. Y. Firdaus, Prof. T. D. Anthopoulos
King Abdullah University of Science and Technology (KAUST)
KAUST Solar Center (KSC)
Division of Physical Sciences and Engineering
Thuwal 23955–6900, Saudi Arabia

J. J. Rech, Prof. W. You
Department of Chemistry
University of North Carolina at Chapel Hill
Chapel Hill, NC 27599, USA

R. Booth, Prof. B. T. O'Connor
Department of Mechanical and Aerospace Engineering, and Organic and Carbon Electronics Lab (ORaCEL)
North Carolina State University
Raleigh, NC 27695, USA

Keywords: organic photovoltaic, tandem solar cells, interconnecting layer

Tandem structure provides a practical way to realize high efficiency organic photovoltaic cells due to the limited optical absorption in organic semiconductors, it can be used to extend the wavelength coverage of the solar spectrum for light harvesting. The interconnecting layer (ICL) between subcells plays a critical role in the reproducibility and performance of tandem solar cells, yet the processability of the ICL has been a challenge. In this work we report on the fabrication of highly reproducible and efficient tandem solar cells by employing a commercially available material, PEDOT:PSS HTL Solar (HSolar), as the hole transporting material used for

the ICL. Comparing with the conventional PEDOT:PSS Al 4083 (c-PEDOT), HSolar offers a better wettability on the underlying non-fullerene photoactive layers, resulting in better charge extraction properties of the ICL. When FTAZ:IT-M and PTB7-Th:IEICO-4F are used as the front cell and the back cell, a power conversion efficiency (PCE) of 14.7% is achieved in the tandem solar cell. To validate the processability of these tandem solar cells, three other research groups have successfully fabricated tandem devices using the same recipe and the highest PCE obtained is 16.1%. With further development of donor polymers and device optimization, our device simulation results show that a $PCE > 22\%$ can be realized in tandem cells in the near future.

1. Introduction

Organic semiconductors are known to have narrow absorption bands and early organic photovoltaic (OPV) devices using fullerenes as an acceptor typically have power conversion efficiencies less than 10%.^[1] With non-fullerene acceptors extending light absorption for light harvesting, the efficiency of these devices started to take off.^[2-4] With the rapid development of non-fullerene acceptors over the past few years, single-junction organic OPV devices have achieved a high power conversion efficiency (PCE) of over 17%.^[5,6] However the low charge carrier mobilities of organic materials limits the active layer thickness thus limiting efficient sunlight harvesting. A strategy to overcome this problem is to stack two solar cells to enhance the light harvesting in OPV devices. By stacking two subcells with complementary optical absorption which covers the range of 450-1050 nm, a $PCE > 17\%$ has recently been achieved.^[7]

In addition to the choice of complimentary photoactive OPV materials to optimize light harvesting, a key component of a tandem solar cell is the interconnecting layer (ICL). An ICL is a layer between the two subcells where opposite charge carriers extracted recombine, and it plays an important role in determining the performance of the tandem devices.^[8,9] An ideal ICL

must allow efficient charge recombination without a loss in fill factor and open-circuit voltage. Its basic structure is a combination of an ultrathin hole extraction layer (HEL) and electron extraction layer (EEL). In addition, the ICL should be optically transparent, and provide chemical protection for the front subcell against dissolution from the solution processing of the back subcell active layer. The formulation of the ICL should not interfere the layers underneath. These requirements severely limit the choice of ICL, and the realization of high efficiency and reproducible tandem devices based on solution-processed ICL remains challenging. A universal, all-solution-based method compatible with non-fullerene based photoactive layers is badly needed for further development of tandem solar cells.

In a single junction solar cell, it is widely accepted that an inverted device outperforms the conventional device. One reason is that in conventional devices, an acidic c-PEDOT used as the hole transport buffer layer can etch the bottom electrode indium tin oxide (ITO), especially at elevated temperature and under humidity. Another reason is that inverted devices eliminate the use of reactive low work function metals as the top electrode.^[10] However, limited by the choice of ICL materials and their physical properties, there are only a few reports on tandem devices utilizing the inverted device architecture with an all-solution process ICL. Table S1 summarize the tandem solar cells reported in the last two years and corresponding ICL used. Amongst the devices reported, only three reported devices have the inverted tandem architecture having a simple HEL/EEL bilayer ICL.^[7,11-12]

Different HEL and EEL candidates have been proposed as an ICL. For electron extraction layers, several polymer ETLs have been reported. However, these materials show optical absorption in the visible region and reduce the light transmitted to the back cell.^[13-15] On the other hand, for HEL poly(3,4-ethylenedioxythiophene):polystyrene sulfonate (PEDOT:PSS) and metal oxides such as MoO_x, V₂O₅ and WO₃ are used. However, metal oxides used in

interconnecting layers did not result in satisfactory device performance, such that an ultrathin (~ 1 nm) metal layer (Al, Ag or Au) was thermally evaporated on top to facilitate the ohmic contact.^[16-18] This thin metal layer reduces the light transmittance through the ICL. In addition, it is questionable whether nanometer thickness control can be consistently achieved when a device is fabricated on a large area substrate. In theory, PEDOT:PSS is an ideal candidate because it is dispersed in water, which is orthogonal to the solvent used for most organic photoactive layers. However the drawback is that it suffers from the high surface energy of water leading to poor wetting of PEDOT:PSS on the organic photoactive layers. Given the hydrophobic nature of the non-fullerene active layer surface, the wetting problem becomes more severe. To address the wetting problem, the PEDOT:PSS formulation needs to be modified for inverted tandem devices processing. Yang *et. al.* demonstrated that by mixing PEDOT:PSS PH500 with sodium polystyrene sulfonate aqueous solution, the resulting ICL has improved durability against chlorobenzene solvent.^[19] Rasi *et. al.* developed a new formulation of PEDOT:PSS by mixing Al 4083 with *n*-propanol.^[9] With a suitable amount of *n*-propanol, it can form a near azeotropic mixture and ensures solvents evaporate completely during spin-coating. With improved wettability, a uniform lossless HEL can be formed on fullerene-based active layers. There are also several reports on improved wetting of PEDOT:PSS with an addition of a small amount of surfactant. However, UV-light soaking is required to form an ohmic contact, and the process can lead to damages of the active layers.^[20,21] These complicated process steps also leads to questionable process control. In summary, while high efficiency tandem solar cells have been reported in the literature, these results are difficult to reproduce even when processing steps are followed in detail. As a result, the progress made on tandem organic solar cells is rather limited.

In this paper, we present a simple yet highly reproducible tandem solar cells using a new version PEDOT:PSS -- HTL Solar (HSolar), in combination of ZnO nanoparticles as electron transport

layer to form the ICL. All materials are commercially available and used as received. They only require direct spin-coating and a moderate temperature thermal annealing to dry out the solvents. We first demonstrate the good wettability of HSolar on a hydrophobic organic active layer, FTAZ:IT-M, with single junction OPV device. When FTAZ:IT-M and PTB7-Th:IEICO-4F are used as the front cell and the back cell respectively to fabricate the tandem devices, a high PCE of 14.7% was achieved. To demonstrate the high compatibility and reproducibility of this ICL, we shared the recipe with three different laboratories around the world. The resulting tandem cells showed good performance without a significant loss in V_{oc} and fill factor with the highest PCE of 16.1%. Using the state of the art OPV photoactive materials for the tandem cells, our simulation results show that a PCE of 22% is possible.

2. Results and Discussion

2.1 Single-junction devices performance

To fabricate tandem cells, FTAZ:IT-M and PTB7-Th:IEICO-4F were chosen as the photoactive layers for the front cell and back cell respectively. **Figure 1a** shows the chemical structures of the donor polymers FTAZ, PTB7-Th and small acceptor molecules IT-M, IEICO-4F. From the UV-Vis measurements, FTAZ:IT-M film has a strong optical absorption in the range of 400-750 nm while PTB7-Th:IEICO-4F has a strong absorption in the range of 500-1000 nm. These photoactive material combinations are well-established with good device performance and reproducibility.^[22-23] Here, FTAZ:IT-M is used as the photoactive material combination for the front cell and PTB7-Th:IEICO-4F is used as the material combination for the back cell. Before the fabrication of tandem devices, the corresponding single junction solar cells were fabricated separately to verify the device performance. First, we report the fabrication of these tandem cells using HSolar in the So Laboratory. The two BHJ blends show complementary absorption covering from 450 nm to 900 nm and both devices showed a good PCE of about 10%. A high J_{sc} of 20.1 mAcm^{-2} was obtained for the PTB7-Th:IEICO-4F OPV cell due to its broad optical

absorption, and a large open-circuit voltage (V_{oc}) of 0.95 V was obtained for the FTAZ:IT-M cell due to the wide bandgap of the polymer donor. The J-V curves of each device under illumination of AM 1.5G light are presented in Figure 1c. The details of the OPV device performance are summarized in **Table 1**. The external quantum efficiency spectra for both optimized devices are shown in **Figure 1d**. It is worth noting that although the EQE spectra of the two devices overlap in the range of 500-750 nm region, the optimized thin active layer thickness (~100 nm) still transmit a certain amount of light through the front cell to the back subcell, and it is expected a high J_{sc} value will be realized in these tandem devices.

2.2. HSolar as hole extraction layer

To demonstrate HSolar ICL for high performance tandem devices, we first study its wetting on the organic photoactive layer. Here, we deposited three different HTLs (HSolar, c-PEDOT and MoO_x) on the FTAZ:IT-M surface to mimic the situation in an inverted single junction solar cell. To compare the wettability between HSolar and c-PEDOT, contact angles were measured and the results are displayed in **Figure 2(c-e)**. For the c-PEDOT sample, the contact angle of water is over 100 degree, indicating that FTAZ:IT-M surface is very hydrophobic. These results are expected as the formulation of c-PEDOT is aqueous and its contact angle of c-PEDOT is similar to water. In comparison the contact angle of HSolar on FTAZ:IT-M, the contact angle was reduced by more than a half. To ensure similar experiments for comparison, the thicknesses of the PEDOT:PSS layers used in these experiments are the same. The data show good wettability of HSolar on a hydrophobic photoactive layer surface, indicating that it should lead to better device performance than c-PEDOT. **Figure 2(a)** shows the corresponding current–voltage (J–V) characteristics for devices using MoO_x , c-PEDOT and HSolar as three different HELs, and the corresponding device performance details are summarized in **Table 2**. Top HEL using c-PEDOT shows a substantially lower fill factor and short-circuit current than the other two. It should also be noted HSolar functions as good as evaporated MoO_x .

To confirm the better device performance of the HSolar device is due to improved wetting, we study the film quality when it is deposited on a FTAZ:IT-M film. The results clearly show a high density of pinholes present in the c-PEDOT film when it is spin coated on top of FTAZ:IT-M, as confirmed by the optical microscopy images in Figure S1. On the other hand, the HSolar film is completely pinhole free, indicating the film quality is much better than for PEDOT Al 4083. The atomic force microscopy results also show that HSolar on FTAZ:IT-M has a much smoother surface with a surface root mean square roughness of 1.9 nm as shown in Figure S2. These results show the much better wettability of HSolar on the photoactive layer compared to c-PEDOT.

To study how wetting affects the electrical characteristics of the resulting devices, we fabricated FTAZ:IT-M hole-only devices, one having PEDOT:PSS as the top hole injection contact and another one having PEDOT:PSS as the bottom contact with a film. The hole current is measured with the two different PEDOT:PSSs. In the case where HSolar or c-PEDOT is used as the bottom injection contact, the measured hole mobilities obtained are about the same ($\sim 3 \times 10^{-4} \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$) for the two different PEDOT:PSSs. However, when they were used as the top contact, the measured hole mobility for the sample with HSolar is $\sim 3 \times 10^{-4} \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$, while the hole mobility value for the sample with c-PEDOT is only $\sim 1 \times 10^{-4} \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$. The data show that there is no difference in the mobility value whether HSolar is used as the bottom or top contact. However, when c-PEDOT is used, the mobility value of the sample with a top PEDOT:PSS contact is only a quarter of the value of the sample with a bottom PEDOT:PSS contact. These data show that the wettability on hydrophobic non-fullerene active layer surface does strongly affect the interface characteristics, and HSolar should be a good candidate to be used in the ICL for tandem cells.

2.3 Tandem cell simulation and optimization.

For optimized tandem solar cells, it is critical to match the photocurrents in the two subcells. Prior to device fabrication, optical modeling was used to provide a guidance for the optimum thickness of the individual subcells. For optical modeling, the optical constants of all layers are required as the input parameters. Here, the refractive indices and extinction coefficients of all layers were measured by spectroscopic ellipsometry. Using the transfer matrix method and drift-diffusion equation, one can calculate the electric field distribution and hence the photocurrent generated in each subcell. The three-dimension plot in **Figure 4(b)** represents the relationship between the short circuit current density of each subcell and their active layer thickness.

The overlapping region of these two planes represents a boundary of J_{sc} matching of the two subcells. Based on the simulation results, the highest J_{sc} is realized when the thicknesses of the optimized front and rear subcell active layers are around 120 and 100 nm respectively. In addition, the spectral and position dependent simulated charge generation rate is displayed in Figure 4(c). An optimum charge generation rate is realized within the back cell in the NIR region between 750 nm and 950 nm, while the front cell still shows good absorption in the visible region. This plot indicates a good utilization of illuminated sunlight throughout the whole spectrum. In addition, the ICL shows almost no absorption, indicative of its minimal optical loss. The cross-section scanning electron microscopy (SEM) image of the tandem device is displayed in Figure 4(d), where a distinct and continuous ICL layer along with a continuous ZnO layer above is displayed, showing good wetting with the layers with the layers below and above it.

Using the results from optical modeling described above, tandem devices consist of the FTAZ: IT-M front cell and the PTB7-Th:IEICO-4F back cell were fabricated. As shown in Figure 4(e),

the optimized tandem solar cell gave a PCE of 14.7%, with a V_{oc} of 1.65 V, a J_{sc} of 14.6 mA/cm² and a FF of 61%. These experimental results match the optical simulation very well. Note that in the tandem devices the V_{oc} is equal to the sum of the individual V_{oc} values of the subcells. To further analyze the tandem device performance, we measured the external quantum efficiency (EQE) of each subcell in the tandem device. To ensure the accuracy and reliability, light and voltage bias are applied during the EQE measurements as proposed by Gilot *et. al.*^[24] As shown in Figure 4(f), the front subcell absorbs most of the high-energy photons within the range of 380–750 nm, with a maximum EQE reaching ~75% at 570 nm. The rear subcell absorbs most of the low-energy photons, with a strong EQE over 70% in the range of 770–850 nm. The rear subcell still shows around 10–30% EQE in the range of 350–700 nm due to the spectral overlap with PTB7-Th:IEICO-4F in this range. Furthermore, the J_{sc} values obtained by integration of front and back subcell EQE curves are 13.9 mA/cm² and 14.1 mA/cm² respectively, which are in good agreement with the experimental data and suggesting a well-balanced current generation in each subcell.

2.4 Reproducibility and Compatibility of ICL

It is known in the research community that reproducibility of solution-processed tandem solar cells is a challenge even when detail experimental procedures described in the literature are followed. They are limited by not only the complexity of fabrication procedures, but also inconsistent processing of the interconnection layer, which hinders the development of tandem OPV devices. To demonstrate the generality of the ICL process, the So Group shared the ICL process recipe with three different research groups: the Ade and Amassian Groups at North Carolina State University and the Anthopoulos Group at the King Abdullah University of Science and Technology. Details of devices and their corresponding performance are listed in **Table 3**. All tandem devices employed different non-fullerene photoactive layers in the front

subcells to demonstrate the versatility of processing of the HSolar HEL. For devices A, B and D, they all have the same device architecture of ITO/ZnO/active layer 1/PEDOT Hsolar/ZnO/active layer 2/MoOx/Ag. For device C, additional thin conjugated polyelectrolyte PFN-Br was inserted between the ZnO layer and the front cell active layer to improve the electron extraction efficiency.

The box charts in **Figure 5** presents the data points of different tandem devices fabricated in different laboratories, each device was repeated 24-46 times. All devices show a small V_{oc} variation of less than 0.08 V. In particular, for device C and device D the V_{oc} variation is 0.04 V or smaller. Among all tandem devices, solar cell utilizing FTAZ:PBDB-T:IT-M has the highest average J_{sc} of 13.8 mA/cm² due to the better light absorption in the visible region.^[25] J_{sc} and FF show a small deviation which originates mainly from the change of active layer film thickness during spin-coating. Combining these photovoltaic parameters, all devices shows good performance and small PCE variation within 1%. Among these devices, the tandem solar cells using PBDB-T:F-M as the front cell and PTB7-Th:IEICO-4F as the back cell has a PCE of 16.1%. The corresponding J-V curve under AM 1.5G light illumination is shown in **Figure 6(a)**. To compare the performance with other results published in the literature, we calculate the efficiency enhancements of the tandem devices related to the corresponding single junction cells and the results are shown in Figure 6(b).^[7,11,12,14,15,26-39] The enhancement factor is defined as the percentage increase of PCE of the tandem cell from the higher efficiency single-junction OPV device. Comparing with the reported values, device C gives the highest enhancement factor of 53%. Tandem devices with the front cells using FTAZ:IT-M or FTAZ:PBDB-T:IT-M also obtain an enhancement > 40%. The origin of the high PCE enhancement is mainly due to the good J_{sc} matching in the tandem device, along with nearly no V_{oc} and small FF losses. Device D achieve a 23% enhancement only due to the large overlapping of optical absorption between the PBDB-T:ITIC and PTB7-Th IEICO-4F subcells in the 650-750 nm region.

To further demonstrate the versatility of the HSolar process, we further use the same recipe to fabricate triple-junction cells. Homo triple-junction tandem structure was used to overcome the decrease in J_{sc} , FF, and ultimately PCE usually observed in single-junction cells when the thickness of the active layer is increased. The device architecture was ITO/ZnO/active layer/HSolar/ZnO/active layer/HSolar/ZnO/active layer/MoO_x/Ag. As shown in **Figure 7**, we chose PTB7-Th:IEICO-4F as the active layers to demonstrate this concept. Based on the transfer matrix model simulation, the optimum thickness of front, middle and back cell should be around 50 nm, 90 nm and 120 nm respectively. The total subcell active layers thickness is 2.5 time thicker than the optimized single-junction PTB7-Th:IEICO-4F solar cell. It also predicts a good charge generation rate throughout all subcells in the range of 450-1000 nm even for the back subcell. Upon optimization, the final tandem device shows nearly no V_{oc} loss (2.1 V) and a good J_{sc} of 8.9 mA/cm². The final PCE of homo tandem device reached 11.2% which is more than 10% higher than its corresponding single-junction cell (9.9%). Cross-section SEM image show that smooth, distinct and continuous layers were successfully formed during fabrication. It is predicted that quadruple or even higher junction tandem device can be easily constructed by sequential deposition of HSolar/ZnO/active layer.

2.5 Path forward to PCE >22% tandem solar cells

With the recent development of a new non-fullerene acceptor, Y6, several single junction solar cells with a PCE >16% have been reported due to the broadband absorption from 400 nm to 950 nm and the low voltage loss. Considering an efficiency enhancement >40% observed in our tandem devices, it is not unrealistic to expect optimized tandem solar cells utilizing Y6-based BHJ blends as a back cell has a great potential to achieve a PCE beyond 22%. To illustrate such a potential, we simulated a tandem solar cell utilizing FTAZ:IT-M and PM6:Y6 as front

cell and back cell respectively, and the resulting device has a high J_{sc} value of 16 mA/cm². **(Figure 8)** Assuming a $V_{oc} = 0.95 + 0.82 = 1.77$ V and $FF = 70\%$, the resulting PCE is 20%. If the fill factor can be further optimized to 77%, a PCE of 22% is achievable. In addition, if the band edge of back cell can be extended to 1000 nm while maintaining the same V_{oc} and FF , the tandem device PCE beyond 22% is definitely possible. Based on these assumptions, we can expect a rapid progress in the efficiency of organic tandem solar cell in the near future.

3. Conclusion

In summary, a simple yet highly compatible interconnecting layer constructed by HSolar/ZnO in inverted tandem OPV device was presented. HSolar inks enjoys good wettability on non-fullerene active layer surfaces and form superior hole extraction layer in OPV devices. All double-junction tandem devices with different active layers show high reproducibility and efficiencies in several laboratories. Among these tandem devices, an excellent PCE of 16.1% has been achieved. In addition, most of the tandem devices achieved more than 40% enhancement from the single-junction OPV device. We also successfully fabricated homo triple-junction tandem device with substantially higher performance than its single-junction counterpart. This ICL should generally applicable to other photoactive system such as organic-perovskites and organic-lead sulphide quantum dots solar cells. Concerning the rapid development of high efficiency organic solar cell photoactive materials, we encourage readers to adopt these ICLs for optimizing future multi-junction devices.

4. Experimental Section

Materials: Poly[4,8-bis(5-(2-ethylhexyl)thiophen-2-yl)benzo[1,2-b;4,5-b']dithiophene-2,6-diyl-alt-(4-(2-ethylhexyl)-3-fluorothieno[3,4-b]thiophene-)-2-carboxylate-2,6-diyl] (PTB7-Th), 3,9-bis(2-methylene-((3-(1,1-dicyanomethylene)-6/7-methyl)-indanone))-5,5,11,11-tetrakis(4-hexylphenyl)-dithieno[2,3-d:2',3'-d']-s-indaceno[1,2-b:5,6-b']dithiophene (IT-M),

and 2,2'-[[4,4,9,9-Tetrakis(4-hexylphenyl)-4,9-dihydro-s-indaceno[1,2-b:5,6-b']dithiophene-2,7-diyl]bis[[4-[(2-ethylhexyl)oxy]-5,2-thiophenediyl]methyldiyn(5,6-difluoro-3-oxo-1H-indene-2,1(3H)-diylidene)]]bis[propanedinitrile (IEICO-4F) were purchased from 1-Material. PEDOT:PSS HTL Solar and Al 4083 were purchased from Heraeus. ZnO nanoparticle (5.6 w/v% in chloroform) was purchased from infinityPV. Poly[4-(5-(4,8-bis(3-butylonyl)benzo[1,2-b:4,5-b']dithiophen-2-yl)thiophen-2-yl)-2-(2-butyloctyl)-5,6-difluoro-7-(thiophen-2-yl)-2H-benzo[d][1,2,3]triazole] (FTAZ) was synthesized as reported previously.^[40,41] These materials were used as received. ZnO sol-gel was prepared by dissolving 2 g of zinc acetate dihydrate into 18.96 mL 2-methoxyethanol and 543.6 μ L of monoethanolamine.

Single OPV Solar Cell Fabrication: The device structure of single OPV device was ITO/ZnO/active layer/MoO_x/Ag. ITO patterned glass substrate was cleaned by acetone and isopropanol in ultrasonic bath followed by UV-ozone treatment. ZnO sol-gel was then spin coated at 4000 RPM onto the ITO substrate and thermal annealed at 150 °C for 30 min. The details of FTAZ:IT-M, PTB7-Th:IEICO-4F, FTAZ:PBDB-T:IT-M and PBDB-T:F-M active layers fabrication were reported elsewhere.^[22,23,42,43] Finally, 10 nm MoO_x and 100 nm Ag were thermally evaporated under high vacuum.

Tandem OPV Solar Cell Fabrication: The device structure of tandem OPV device was ITO/ZnO/active layer/PEDOT:PSS HTL Solar/ZnO/active layer/MoO_x/Ag. The procedures for front cell and back cell fabrication are the same as single OPV device. For interconnecting layer, PEDOT:PSS (Clevios HTL Solar) was spin coated onto FTAZ:IT-M at 5000 RPM for 60s, following 105 °C thermal annealing for 10 min. After cooling down of substrate, ZnO nanoparticle (5.6% w/v in chloroform) was then spin coated on top at 7000 RPM for 60s, following 105 °C thermal annealing for 10 min. The active areas of OPV devices were covered

by a 4 mm² aperture during characterization. Light intensity was calibrated using a KG-5 reference cell. The values of mismatch factor for both subcells under the solar simulator were calculated according to ref. 44, and the values were ~1.

Supporting Information

Supporting Information is available from the author.

Acknowledgements

This work was supported by the NextGen Nano Limited, Office of Naval Research Grant N00014-17-1-2242 and N00014-17-1-2204, National Science Foundation Award CBET-1639429, and the North Carolina State University.

References

- [1] Y. He, Y. Li, *Phys. Chem. Chem. Phys.* **2011**, *13*, 1970.
- [2] C. Yan, S. Barlow, Z. Wang, H. Yan, A. K.-Y. Jen, S. R. Marder, X. Zhan, *Nat. Rev. Mater.* **2018**, *3*, 18003.
- [3] J. Hou, O. Inganäs, R. H. Friend, F. Gao, *Nat. Mater.* **2018**, *17*, 119.
- [4] P. Cheng, G. Li, X. Zhan, Y. Yang, *Nat. Photonics* **2018**, *12*, 131.
- [5] Y. Lin, B. Adilbekova, Y. Firdaus, E. Yengel, H. Faber, M. Sajjad, X. Zheng, E. Yarali, A. Seitkhan, O. M. Bakr, A. El-Labban, U. Schwingenschlögl, V. Tung, I. McCulloch, F. Laquai, T. D. Anthopoulos, *Adv. Mater.* **2019**, *31*, 1902965.
- [6] Q. Liu, Y. Jiang, K. Jin, J. Qin, J. Xu, W. Li, J. Xiong, J. Liu, Z. Xiao, K. Sun, S. Yang, X. Zhang, L. Ding, *Science Bulletin*, **2020**.
- [7] L. Meng, Y. Zhang, X. Wan, C. Li, X. Zhang, Y. Wang, X. Ke, Z. Xiao, L. Ding, R. Xia, H.-L. Yip, Y. Cao, Y. Chen, *Science* **2018**, *361*, 1094.
- [8] S. Lu, D. Ouyang, W. C. H. Choy, *Sci. China: Chem.* **2017**, *60*, 460.
- [9] D. D. C. Rasi, K. H. Hendriks, G. H. L. Heintges, G. Simone, G. H. Gelinck, V. S. Gevaerts, R. Andriessen, G. Pirotte, W. Maes, W. Li, M. M. Wienk, R. A. J. Janssen, *Solar RRL* **2018**, *2*, 1800018.
- [10] K. Wang, C. Liu, T. Meng, C. Yi, X. Gong, *Chem. Soc. Rev.* **2016**, *45*, 2937.
- [11] L. Meng, Y.-Q.-Q. Yi, X. Wan, Y. Zhang, X. Ke, B. Kan, Y. Wang, R. Xia, H.-L. Yip, C. Li, Y. Chen, *Adv. Mater.* **2019**, *31*, 1804723.
- [12] S. Chen, G. Zhang, J. Liu, H. Yao, J. Zhang, T. Ma, Z. Li, He Yan, *Adv. Mater.* **2017**, *29*, 1604231.
- [13] Z. Wu, C. Sun, S. Dong, X.-F. Jiang, S. Wu, H. Wu, H.-L. Yip, F. Huang, Y. Cao, *J. Am. Chem. Soc.* **2016**, *138*, 2004.
- [14] G. Liu, J. Jia, K. Zhang, X. Jia, Q. Yin, W. Zhong, L. Li, F. Huang, Y. Cao, *Adv. Energy Mater.* **2019**, *9*, 1803657.
- [15] K. Zhang, R. Xia, B. Fan, X. Liu, Z. Wang, S. Dong, H.-L. Yip, L. Ying, F. Huang, Y. Cao, *Adv. Mater.* **2018**, *30*, 1803166.
- [16] T. T. Larsen-Olsen, E. Bundgaard, K. O. Sylvester-Hvid, F. C. Krebs, *Org. Electron.* **2011**, *12*, 364.
- [17] C. H. Chou, W. L. Kwan, Z. R. Hong, L. M. Chen, Y. Yang, *Adv. Mater.* **2011**, *23*, 1282.
- [18] W. Liu, S. Li, J. Huang, S. Yang, J. Chen, L. Zuo, M. Shi, X. Zhan, C.-Z. Li, H. Chen, *Adv. Mater.* **2016**, *28*, 9729.
- [19] J. Yang, R. Zhu, Z. Hong, Y. He, A. Kumar, Y. Li, Y. Yang, *Adv. Mater.* **2011**, *23*, 3465.

- [20] J. Adams, G. D. Spyropoulos, M. Salvador, N. Li, S. Strohm, L. Lucera, S. Langner, F. Machui, H. Zhang, T. Ameri, M. M. Voigt, F. C. Krebs, C. J. Brabec, *Energy Environ. Sci.* **2015**, 8, 169.
- [21] S. Kouijzer, S. Esiner, C. H. Frijters, M. Turbiez, M. M. Wienk, R. A. J. Janssen, *Adv. Energy Mater.* **2012**, 2, 945.
- [22] L. Ye, Y. Xiong, Q. Zhang, S. Li, C. Wang, Z. Jiang, J. Hou, W. You, H. Ade, *Adv. Energy Mater.* **2018**, 30, 1705485.
- [23] X. Song, N. Gasparini, L. Ye, H. Yao, J. Hou, H. Ade, D. Baran, *ACS Energy Lett.* **2018**, 3, 669.
- [24] J. Gilot, M. M. Wienk, R. A. J. Janssen, *Adv. Funct. Mater.* **2010**, 20, 3904.
- [25] H. Hu, L. Ye, M. Ghasemi, N. Balar, J. J. Rech, S. J. Stuard, W. You, B. T. O'Connor, H. Ade, *Adv. Mater.* **2019**, 31, 1808279.
- [26] X. Che, Y. Li, Y. Qu, S. R. Forrest, *Nat. Energy* **2018**, 3, 422.
- [27] F. X. Chen, J. Q. Xu, Z. X. Liu, M. Chen, R. Xia, Y. Yang, T. K. Lau, Y. Zhang, X. Lu, H. L. Yip, A. K. Jen, H. Chen, C.-Z. Li, *Adv. Mater.* **2018**, 30, 1803769.
- [28] B. Guo, W. Li, G. Luo, X. Guo, H. Yao, M. Zhang, J. Hou, Y. Li, W.-Y. Wong, *ACS Energy Lett.* **2018**, 3, 2566.
- [29] Z. P. Yu, Z. X. Liu, F. X. Chen, R. Qin, T. K. Lau, J. L. Yin, X. Kong, X. Lu, M. Shi, C. Z. Li, H. Chen, *Nat. Commun.* **2019**, 10, 2152.
- [30] Y. Zhang, B. Kan, Y. Sun, Y. Wang, R. Xia, X. Ke, Y. Yi, C. Li, H. Yip, X. Wan, Y. Cao, Y. Chen, *Adv. Mater.* **2018**, 30, 1707508.
- [31] K. Zhang, B. Fan, R. Xia, X. Liu, Z. Hu, H. Gu, S. Liu, H. L. Yip, L. Ying, F. Huang, Y. Cao, *Adv. Energy Mater.* **2018**, 8, 1703180.
- [32] W. Huang, S.-Y. Chang, P. Cheng, D. Meng, B. Zhu, S. Nuryyeva, C. Zhu, L. Huo, Z. Wang, M. Wang, Y. Yang, *Nano Lett.* **2018**, 18, 7977.
- [33] D. D. C. Rasi, P. M. J. G. van Thiel, H. Bin, K. H. Hendriks, G. H. L. Heintges, M. M. Wienk, T. Becker, Y. Li, T. Riedl, R. A. J. Janssen, *Solar RRL* **2019**, 3, 1800366.
- [34] S. Lu, H. Lin, S. Q. Zhang, J. H. Hou, W. C. H. Choy, *Adv. Energy Mater.* **2017**, 21, 1701164.
- [35] S.-Y. Chang, Yu.-C. Lin, P. Sun, Y.-T. Hsieh, L. Meng, S.-H. Bae, Y.-W. Su, W. Huang, C. Zhu, G. Li, K.-H. Wei, Y. Yang, *Solar RRL* **2017**, 1, 1700139.
- [36] Z. Shi, Y. Bai, X. Chen, R. Zeng, Z. Tan, *Sustainable Energy Fuels* **2019**, 3, 910.
- [37] J. Yuan, M. J. Ford, Y. Xu, Y. Zhang, G. C. Bazan, W. Ma, *Adv. Energy Mater.* **2018**, 8, 1703291.
- [38] Z. Shi, H. Liu, L. Xia, Y. Bai, F. Wang, B. Zhang, T. Hayat, A. Alsaedi, Z. Tan, *Chin. J. Chem.* **2018**, 36, 194.
- [39] T. Becker, S. Trost, A. Behrendt, I. Shutsko, A. Polywka, P. Görrn, P. Reckers, C. Das, T. Mayer, D. Di Carlo Rasi, K. H. Hendriks, M. M. Wienk, R. A. J. Janssen, T. Riedl, *Adv. Energy Mater.* **2018**, 8, 1702533.
- [40] S. C. Price, A. C. Stuart, L. Yang, H. Zhou, Wei You, *J. Am. Chem. Soc.* **2011**, 133, 4625.
- [41] Q. Zhang, L. Yan, X. Jiao, Z. Peng, S. Liu, J. J. Rech, E. Klump H. Ade, F. So, W. You, *Chem. Mater.* **2017**, 29, 5990.
- [42] H. Hu, L. Ye, M. Ghasemi, N. Balar, J. J. Rech, S. J. Stuard, W. You, B. T. O'Connor, H. Ade, *Adv. Mater.* **2019**, 3, 1808279.
- [43] Y. Zhang, B. Kan, Y. Sun, Y. Wang, R. Xia, X. Ke, Y.-Q.-Q. Yi, C. Li, H. - L. Yip, X. Wan, Y. Cao, Y. Chen, *Adv. Mater.* **2018**, 30, 1707508.
- [44] V. Shrotriya, G. Li, Y. Yao, T. Moriarty, K. Emery, Y. Yang, *Adv. Funct. Mater.* **2018**, 16, 2016.

Figure 1. (a) Chemical structure of active layer materials. (b) Optical absorption of FTAZ:IT-M and PTB7-Th:IEICO-4F thin films. (c) Corresponding J-V characteristics under AM1.5G solar spectrum. (d) External quantum efficiency spectrum of single cells.

Table 1. Device performance of single-junction inverted OPV cells.

Active layer	J_{sc} (mA cm ⁻²)	V_{oc} (V)	FF (%)	PCE (%)
FTAZ : IT-M	16.3	0.95	64	9.8
PTB7-Th : IEICO-4F	20.1	0.73	68	9.9

Figure 2. (a) Schematic diagram of single-junction FTAZ : IT-M solar cell using different hole extraction layers, (b) their corresponding J-V characteristics under AM1.5G solar spectrum. (c-e) Contact angle of water, c-PEDOT and HSolar on FTAZ : IT-M thin film surface.

Table 2: FTAZ : IT-M OPV devices performance with different hole transporting layers.

HEL used	J_{sc} (mA cm ⁻²)	V_{oc} (V)	FF (%)	PCE (%)
MoO _x	16.3	0.95	64	9.8
c-PEDOT	15.1	0.95	53	7.6
HSolar	16.6	0.95	63	9.9

Figure 3. J-V characteristics of hole-only devices with different hole transport layers (a) underneath or (b) on top of FTAZ : IT-M layer..

Figure 4. (a) Layout of the double-junction tandem device structure (b) Simulated J_{sc} in each subcells as a function of subcell active layer thicknesses. The dash line in intersection of two planes illustrates a balanced J_{sc} between front cell and back cell. (c) Simulated relative charge generation rate distribution (d) cross-section SEM image of the tandem device. (e) Corresponding J-V characteristics under AM1.5G solar spectrum. (f) EQE of the subcells.

Figure 5. Box charts of (a) J_{sc} , (b) FF, (c) V_{oc} and (d) PCE of different tandem OPV devices fabricated in different laboratories. Total number of each devices are listed in Table 3.

Table 3. Details of tandem device structures fabricated in different laboratories. Performances of their corresponding single cell are listed in Table S5.

Device	Front cell	Back cell	J_{sc} (mA/cm ²)	V_{oc} (V)	FF (%)	PCE (%)	Number of devices
A	FTAZ:IT-M	PTB7-Th:IEICO-4F	13.3 ± 0.28	1.6 ± 0.02	65 ± 0.82	13.9 ± 0.26	46
B	FTAZ:PBDB-T:IT-M	PTB7-Th:IEICO-4F	13.8 ± 0.39	1.54 ± 0.02	65 ± 1.02	13.8 ± 0.26	24
C	PBDB-T:F-M	PTB7-Th:IEICO-4F	13.4 ± 0.37	1.66 ± 0.01	70 ± 1.44	15.6 ± 0.25	30
D	PBDB-T:ITIC	PTB7-Th:IEICO-4F	12.0 ± 0.28	1.49 ± 0.01	64 ± 0.99	11.4 ± 0.23	45

Figure 6 (a) J-V characteristics of tandem devices utilizing PBDB-T:ITIC front cell and PTB7-Th:IEICO-4F back cell under AM1.5G solar spectrum. (b) Comparison of efficiency enhancement from single subcells, as a function of tandem device efficiency.

Figure 7. (a) Layout of the triple-junction tandem device structure (b) Simulated J_{sc} in each subcells as a function of subcell active layer thicknesses. (c) Simulated relative charge generation rate distribution (d) cross-section SEM image of the triple-junction tandem device. (e) Corresponding J-V characteristics under AM1.5G solar spectrum.

Table 4: PTB7-Th : IEICO-4F OPV devices performance with different structures.

Device structure	J_{sc} (mA cm ⁻²)	V_{oc} (V)	FF (%)	PCE (%)
Single junction	20.1	0.73	68	9.9
Triple junction	8.9	2.10	60	11.2

Figure 8. (a) Simulated subcell thickness dependent J_{sc} in a tandem device with FTAZ:IT-M and PM6:Y6 as front cell and back cell respectively. (b) Simulated subcell thickness dependent J_{sc} with back cell active layer absorption edge extended to 1000 nm.

Supporting Information

High Performance Tandem Organic Solar Cells using HSolar as the Interconnecting Layer

Carr Hoi Yi Ho, Taesoo Kim, Yuan Xiong, Yuliar Firdaus, Xueping Yi, Qi Dong, Jeromy J. Rech, Abay Gadisa, Ronald Booth, Brendan T. O'Connor, Aram Amassian, Harald Ade, Wei You, Thomas D. Anthopoulos, Franky So**

Table S1: Tandem OPV devices reported in the last two years and corresponding ICL used

Front subcell	ITL	Back subcell	J _{sc} (mA/cm ²)	V _{oc} (V)	FF (%)	PCE (%)	Enhancement (%)	Ref.
PBDB-T:F-M	m-PEDOT/ZnO	PTB7-Th:O6F-4F:PC71BM	14.4	1.64	73	17.3	30	1
DTDCPB:C ₇₀	BPhen:C60/Ag/PEDOT:PSS	PTB7-Th:BT-CIC	13.3	1.59	71	15.0	40	2
PBDB-T-2F:TfIF-4FIC	PF3N-2TNDI/Ag/PEDOT:PSS	PTB7-Th:PCDTBT:IEICO-4F	13.6	1.60	69	15.0	22	3
PBDB-T-2F:TfIF-4FIC	PF3N-2TNDI/Ag/PEDOT:PSS	PTB7-Th:PCDTBT:IEICO-4F	13.6	1.60	69	14.8	20	3
PBDB-T:ITIC	MoOx/Ag/ZnO/PFN-Br	PTB7-Th:T2	13.2	1.56	71	14.6	35	4
PBDB-T-2F:F-M	m-PEDOT/ZnO	PBDB-T-2F:NNBDT	10.7	1.82	75	14.5	22	5
PBD1:PC ₇₁ BM	ZnO/n-PEDOT:PSS	PTB7-Th:IEICO-4F	12.3	1.61	72	14.2	39	6
PBDB-TF:PTIC	MoOx/Ag/ZnO/PFN-Br	PTB7-Th:IEICO-4F	12.8	1.61	68	14.0	36	7
PBDB-T:F-M	ZnO/n-PEDOT:PSS	PTB7-Th: NOBDT	11.7	1.71	70	13.9	36	8
PSTzBI-EHp:PC ₇₁ BM	PNDIT-F3N:PEI/PEDOT:PSS	PBDTTT-E-T:IEICO	11.4	1.72	67	12.9	34	9
PDBT-T1:TPH-Se:ITIC	MoOx/m-PEDOT:PSS/ZnO	PTB7-Th:IEICO-4F	7.8	2.52	63	12.5	9	10
PTzBI-Si:N2200	PNDIT-F3N:PEI/PEDOT:PSS	PTzBI-Si:N2200	8.9	1.72	72	10.9	9	11
P3TEA:SF-PDI2	m-PEDOT:PSS/ZnO	P3TEA:SF-PDI2	8.3	2.13	61	10.8	14	12
J71:ITIC	SnOx/PEDOT:PSS	PMDPP3T:PC60BM	11.1	1.48	63	10.4	14	13
PTB7:PC ₇₁ BM	PMA/M-PEDOT/ZnO	PDPP4T-2F:PC70BM	10.5	1.47	67	10.3	24	14
PBDTTBO:PC71BM	MoOx/PEDOT:PSS/Zr-Acac	PDTP-DFBT:PC71BM	11.5	1.48	57	9.8	36	15
PBDTBDD:PC70BM	MoOx/Ag/PFN-Br	PTB7-Th:IEICO-4F	11.5	1.50	56	9.7	28	16
PTP8:p(NDI2HD-T)	MoOx/Ag/ZnO	PBFSF:N2200	8.0	1.77	59	8.3	36	17
PTB7-Th:PC ₇₁ BM	MoO ₃ /Ag/TIPD	PTB7-Th:PC71BM	8.8	1.53	60	8.1	12	18
PCDTBT:PC ₇₁ BM	MoOx/SnOx	PDPP3T:PC60BM	8.4	1.48	61	7.6	41	19
<i>PBDB-T:F-M</i>	<i>PEDOT HSolar/ZnO</i>	<i>PTB7-Th:IEICO-4F</i>	<i>14.6</i>	<i>1.64</i>	<i>67</i>	<i>16.1</i>	<i>53</i>	<i>This work</i>
<i>FTAZ:IT-M</i>	<i>PEDOT HSolar/ZnO</i>	<i>PTB7-Th:IEICO-4F</i>	<i>14.6</i>	<i>1.65</i>	<i>61</i>	<i>14.7</i>	<i>48</i>	<i>This work</i>
<i>FTAZ:PBDB-T:IT-M</i>	<i>PEDOT HSolar/ZnO</i>	<i>PTB7-Th:IEICO-4F</i>	<i>14.6</i>	<i>1.58</i>	<i>66</i>	<i>14.5</i>	<i>46</i>	<i>This work</i>
<i>PBDB-T:ITIC</i>	<i>PEDOT HSolar/ZnO</i>	<i>PTB7-Th:IEICO-5F</i>	<i>12.7</i>	<i>1.5</i>	<i>63</i>	<i>12</i>	<i>23</i>	<i>This work</i>

Table S2. Specification for the PEDOT:PSS - HTL Solar. Adopted from Ref. 20.

Resistivity	1 - 10 Ω .cm
Solid content	1.0 - 1.3 wt.% (in water)
Viscosity	8 to 30 mPa.s
PEDOT:PSS ratio	1:2.5
Work Function	4.8 - 5.0 eV

Figure S1. Transmittance spectrum of HSolar and ZnO thin film.

Figure S2. 200 μm × 200 μm microscopy images of different layer stacks.

Figure S3. Atomic force microscopy images of different layer stacks.

Table S3. Surface roughness of different layer stacks obtained from Figure S3.

Layers	R_q (nm)
FTAZ : IT-M	0.596
FTAZ : IT-M / c-PEDOT	4.571
FTAZ : IT-M / HSolar	1.920
FTAZ : IT-M / c-PEDOT/ ZnO	1.919
FTAZ : IT-M / HSolar / ZnO	0.901

Figure S4. The (a) extinction coefficients and (b) refractive indices of FTAZ:IT-M and PTB7-Th: IEICO-4F films used for device simulation.

Figure S5. Energy level diagram of the tandem solar cell.

Table S4. Optimization of ICLs in FTAZ:IT-M/PTB7-Th:IEICO-4F tandem devices.

HSolar/ZnO thickness (nm)	J_{sc} (mA/cm ²)	V_{oc} (V)	FF (%)	PCE (%)
20/40	14.0	1.65	61	14.1
40/100	14.3	1.64	60	14.1
20/100	14.6	1.65	61	14.7
20/150	14.0	1.64	58	13.3

Table S5. Photovoltaic parameters of each subcells in tandem device B, C and D.

Active layer	J_{sc} (mA/cm ²)	V_{oc} (V)	FF (%)	PCE (%)
FTAZ:PBDB-T:IT-M	18.10	0.95	74	12.7
PBDB-T:F-M	16.04	1.0	65	10.4
PBDB-T:ITIC	15.58	0.86	66	8.7

References

- [1] L. Meng, Y. Zhang, X. Wan, C. Li, X. Zhang, Y. Wang, X. Ke, Z. Xiao, L. Ding, R. Xia, H.-L. Yip, Y. Cao, Y. Chen, *Science* **2018**, *361*, 1094.
- [2] X. Che, Y. Li, Y. Qu, S. R. Forrest, *Nat. Energy* **2018**, *3*, 422.
- [3] G. Liu, J. Jia, K. Zhang, X. Jia, Q. Yin, W. Zhong, L. Li, F. Huang, Y. Cao, *Adv. Energy Mater.* **2019**, *9*, 1803657.
- [4] F. X. Chen, J. Q. Xu, Z. X. Liu, M. Chen, R. Xia, Y. Yang, T. K. Lau, Y. Zhang, X. Lu, H. L. Yip, A. K. Jen, H. Chen, C.-Z. Li, *Adv. Mater.* **2018**, *30*, 1803769.
- [5] L. Meng, Y.-Q.-Q. Yi, X. Wan, Y. Zhang, X. Ke, B. Kan, Y. Wang, R. Xia, H.-L. Yip, C. Li, Y. Chen, *Adv. Mater.* **2019**, *31*, 1804723.
- [6] B. Guo, W. Li, G. Luo, X. Guo, H. Yao, M. Zhang, J. Hou, Y. Li, W.-Y. Wong, *ACS Energy Lett.* **2018**, *3*, 2566.
- [7] Z. P. Yu, Z. X. Liu, F. X. Chen, R. Qin, T. K. Lau, J. L. Yin, X. Kong, X. Lu, M. Shi, C. Z. Li, H. Chen, *Nat. Commun.* **2019**, *10*, 2152.
- [8] Y. Zhang, B. Kan, Y. Sun, Y. Wang, R. Xia, X. Ke, Y. Yi, C. Li, H. Yip, X. Wan, Y. Cao, Y. Chen, *Adv. Mater.* **2018**, *30*, 1707508.
- [9] K. Zhang, B. Fan, R. Xia, X. Liu, Z. Hu, H. Gu, S. Liu, H. L. Yip, L. Ying, F. Huang, Y. Cao, *Adv. Energy Mater.* **2018**, *8*, 1703180.
- [10] W. Huang, S.-Y. Chang, P. Cheng, D. Meng, B. Zhu, S. Nuryyeva, C. Zhu, L. Huo, Z. Wang, M. Wang, Y. Yang, *Nano Lett.* **2018**, *18*, 7977.
- [11] K. Zhang, R. Xia, B. Fan, X. Liu, Z. Wang, S. Dong, H.-L. Yip, L. Ying, F. Huang, Y. Cao, *Adv. Mater.* **2018**, *30*, 1803166.
- [12] S. Chen, G. Zhang, J. Liu, H. Yao, J. Zhang, T. Ma, Z. Li, He Yan, *Adv. Mater.* **2017**, *29*, 1604231.
- [13] D. D. C. Rasi, P. M. J. G. van Thiel, H. Bin, K. H. Hendriks, G. H. L. Heintges, M. M. Wienk, T. Becker, Y. Li, T. Riedl, R. A. J. Janssen, *Solar RRL* **2019**, *3*, 1800366.
- [14] S. Lu, H. Lin, S. Q. Zhang, J. H. Hou, W. C. H. Choy, *Adv. Energy Mater.* **2017**, *21*, 1701164.
- [15] S.-Y. Chang, Yu.-C. Lin, P. Sun, Y.-T. Hsieh, L. Meng, S.-H. Bae, Y.-W. Su, W. Huang, C. Zhu, G. Li, K.-H. Wei, Y. Yang, *Solar RRL* **2017**, *1*, 1700139.
- [16] Z. Shi, Y. Bai, X. Chen, R. Zeng, Z. Tan, *Sustainable Energy Fuels* **2019**, *3*, 910.
- [17] J. Yuan, M. J. Ford, Y. Xu, Y. Zhang, G. C. Bazan, W. Ma, *Adv. Energy Mater.* **2018**, *8*, 1703291.
- [18] Z. Shi, H. Liu, L. Xia, Y. Bai, F. Wang, B. Zhang, T. Hayat, A. Alsaedi, Z. Tan, *Chin. J. Chem.* **2018**, *36*, 194.
- [19] T. Becker, S. Trost, A. Behrendt, I. Shutsko, A. Polywka, P. Görrn, P. Reckers, C. Das, T. Mayer, D. Di Carlo Rasi, K. H. Hendriks, M. M. Wienk, R. A. J. Janssen, T. Riedl, *Adv. Energy Mater.* **2018**, *8*, 1702533.
- [20] <https://www.ossila.com/products/pedot-pss?variant=30366225301600>